CRITICALITY CONDITIONS AND SUSTENANCE OF REACTION IN NATURALLY CONSTITUTED NUCLEAR REACTORS

By

NAVJYOTI SINGH

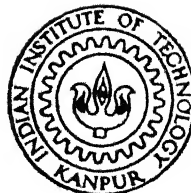**NUCLEAR ENGINEERING AND TECHNOLOGY PROGRAMME**

INDIAN INSTITUTE OF TECHNOLOGY, KANPUR

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CRITICALITY CONDITIONS AND SUSTENANCE OF REACTION IN NATURALLY CONSTITUTED NUCLEAR REACTORS

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By
NAVJYOTI SINGH

to the
NUCLEAR ENGINEERING AND TECHNOLOGY PROGRAMME
INDIAN INSTITUTE OF TECHNOLOGY, KANPUR
DECEMBER, 1981

DEDICATED

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MY FAMILY

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Certificate

Certified that the work presented in this thesis entitled " Criticality Conditions and Sustenance of Reaction in Naturally constituted Nuclear Reactors " by Navjyoti Singh, has been done under my supervision and that it has not been submitted elsewhere.

2nd December, 1981.

Dr. K. SRI RAM
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Kanpur.

ACKNOWLEDGEMENT

It was fortunate that I came across this topic which is close to my sensibility otherwise I would have done something quite remote with uninterestedness of compulsion for my M.Tech. I don't know who is to be thanked for this. It was just a chance.

I thank my teacher Prof. K. Sri Ram for guiding me through this venture. Also I acknowledge my indebtness to Ravichandran and R.K. Patnaik who initiated me into an art of computation. Something vital would have been less without constant encouragement and inspiration by Naik, Keshav Ganguly Muddhu Krishnan, Harpreet, S.B. Kulkarni, Sanjeev and many other friends and wellwishers.

Finally, I thank Mr. J.P. Gupta who laboured patiently in preparing typescript and R.S. Tripathi whose neat cyclo-styling helped in producing these copies.

NAVJYOTI SINGH

ABSTRACT

To study criticality condition of Natural reactors of Oklo a computational model, which describes neutron balance in ore, is developed using thermal neutron diffusion equation for homogenious medium. Criticality equation is modified to includ contribution from epithermal fission of U-235. Also included are temperature dependence of various quantities. Main paramet determining criticality are discussed in the background of geo-chemical information available about reactors. It is observed that moderation feature of Oklo rocks were as good as heavy wate and neutron leakage was much less compared to man-made reactors.

To study sustenance of chain reaction once criticality was achieved, burnup/buildup equations of twelve important isotopes is solved by numerical integration scheme. In most favourable situations reaction would have stopped after 10^{16} n/c to 10^{19} n/cm^2 fluence. To sustain it further, hypothesis of continued mineralisation was examined for various criticality situations. Quantity of additional mineralisation needed is obtained for fluence to reach 10^{21} n/cm^2 It was concluded that either average fluence reached was 5×10^{20} n/cm^2 or if it was not so hypothesis of accretion of uranium during reactor operation alone can not explain sustenance of reaction.

CONTENT

LIST OF SYMBOLS

B^2 Geometric Buckling of reactors

d_r Density of rock

d_u Partial density of uranium in rock

d_w Density of water in rock pores

D_{th} Thermal diffusion coefficient

f Thermal utilisation factor

I_{eff} Effective resonance integral

$I_{inf.dil.}$ Infinite dilution resonance integral

K_{eff} Effective neutron multiplication factor

K_{inf} Infinite neutron multiplication factor

k Boltzmann constant

L^2 Diffusion area

m weight of loose water per unit weight of UO_2

m' Content of water chemically bound to gangue per unit gangue

          Number density of ith isotope per unit volume

          Non-leakage probability for i^{th} isotope

          Spectrum index

          Reactor operation time

          Temperature of melieu

          Neutron temperature

μ	Lethargy
U%	Percentage of uranium in rock by weight
V%	Volume content of loose water in rock in percentage
x	B^{10} equivalent content of poison in parts per million uranium atoms
X	Weight of gangue per unit weight of UO_2 in rock
Y	Weight of water per unit weight of UO_2 in rock
β	Factor which takes care of epithermal fission absorption of U-235
γ_i^j	Yield of jth species from fission of ith isotope
η	Regeneration factor
ε	Fast fission factor
λ_i	Radioactive decay constant of ith isotope
φ	Flux of neutrons
θ	Fluence
$\bar{\nu}_i$	Number of neutrons born per fission of ith isotope
$\bar{\mu}_i$	Average of cosine of scattering angle of ith isotope
$\bar{\xi}$	Average logarithmic energy decrement in slowing down, For i^{th} species ξ_i is average logarithmic energy decrement
τ	Geological time in billion years.
τ'	Neutron Age of slowing down
σ_j^i	Microscopic thermal cross-section of ith isotope for j^{th} process. j is a,f,s or t for absorption, fission, scattering and total.

$\sigma^i_{s/e}$ Microscopic epithermal scattering cross-sections for i^{th} isotope

Σ_i Macroscopic thermal cross-section of jth process. j can be a,s or t for absorption scattering or total.

$\Sigma_{s/e}$ Macroscopic epithermal scattering cross-section

Once upon a time some 1.8 billion years back there worked atleast six nuclear reactors underneath one quite corner of earth which today is called by the name of Oklo. They worked at the time when earth was inhabited not by men not by fishes nor even by plants but by numerous tiny creatures, virus and bacteria. One of the bacterial species called blue-green algee had by then turned out victorious in its promissiv struggle for transforming whole of earth making its atmosphere oxygenic so that in future plants, animal and men could inhabi tate earth. It was their labor that had made possible springi up of these reactors, though it was unintended (see page 15).

Not before June, 1972 had man known about these little wonders. P.K. Kuroda says " Prior to this man believed that world's first nuclear chain reaction was artificial and had occured at the University of Chicago on December 2, 1942. The discovery of the Oklo phenomenon thus contradicted this view, since it contained experimental evidence that nature, not humans, had produced the first chain reaction."[1]. This is from a man who predicted such occurances 16 years prior to the discovery of Oklo [2,3]. The story of discovery itself is a unique one. On June, 1972, a team of the French Atomic Energy Commission, Banzigue et al. [4], were controlling on a routine basis the input of uranium at the French uranium gas diffusion Plant of Pierrelate when a sample of secondary uranium showed U-235 content of $0.7171 \pm 0.001\%$, which is

slightly different from its thought to be constant value of
0.7202 $\pm$ 0.0006% on earth. At first it was thought that
this minor deviation is because of experimental error but
repeated experiments led them to rule out this possibility
and suspect contamination by commercial depleted uranium in
supplied uranium. Search for the source of contamination
began, it had been supplied by French Company, COMUF, opera-
ting two mines of Oklo and Mounana in Gabon. Soon it was
known that uranium traded by this company for one and a half
year, amounting to 700 tons, had less than normal U-235
concentration. On examination of mineral of Oklo to their
surprise ore itself was found depleted in U-235. In one rock
sample depletion was upto 0.441% . Various hypothesis were
advanced to account for this anamoly. If it was because of
nuclear chain reaction their would be abundance of fission
products. Soon rare-earth fission products (Nd) in large
quantities were detected by mass spectrometry. This was a
decisive evidence for nuclear chain reaction and in September,
1972, Neuilly et al. [5] confidently announced that Oklo
deposits had been the site of Natural chain reaction. By
March, 1973 CEA formalised investigation of Oklo phenomenon
as " Project Francevelle" under leadership of Dr. R. Neudet [6]
thus initiating on site examination of mine. In the beginning
this team identified three distinct uranium rich zones in
rocks where marked depletion was found. One record sample
contained surprisingly low value of 0.296% U-235. One sample
even showed slight enrichment with U-235 content of 0.74% .

Fortunately good correlation between U-235 isotopic abundance, uranium abundance and fission products abundance was establish(indicating that reactor fossile had remained intact after reaction stopped in it. Rocks of Oklo had kept complete record of a unique event. Continued investigation soon led to identi- fication of six fossilised reaction zones in same site. At tha time in a reflective mood Neudet wrote "we don't know which should be considered the more astonishing, that these (reactor) mechanisms existed or that we find such detailed evidence nearly two billion years later" [6]. Geological age of working of these reactors by that time had been estimated to be close to 2 billion years.

Since then two international meetings on Oklo phenomenon have been held with patronage of IAEA at Libreville in June, 1975 [7] and in Paris in September, 1977 [8]. Wide range of extensive multidisciplinary studies were reported in these conferences ranging from geological, geochemical, radiological, patrological to reactor physics aspect of Oklo phenomenon. These studies have illustrated how Oklo phenomenon challenges over understanding of wide variety of complex problems like ageing of geological and geochemical freatures over a long time scale, stability of radiation damage features in insulators, radiation induced corrosion, composition and circulation of deep ground water, redistribution and stability of elements in nature over a very long time scale. Also Oklo phenomenon is an challenge to analytic methods used in variety of fields like earth sciences, irradiation effects in solids, radio-

chemistry and so forth. Only the most refined methods used
in geochronology, rare-gas determinations and ion-microprobe
analysis can be exploited in the "non-closed" geochemical
system of the Oklo reactors, where in addition to other processe
neutron fluences have been active to further modify isotopic
abundances. Besides these problems reactor physics aspect of
these reactors is challenging. Problem of understanding
sustenance of reaction, operating mode of reactors and duration
of reaction are still open.

One of the discovery of far reaching consequences from
Oklo has been a finding that quartz variety found in Oklo
ore has the highest track retentivity ever reported for an
insulator [9]. Laboratory experiments show that fission track
registered in the Oklo quartz should be stable at $300\,^{\circ}C$, over
a time scale of 10 billion years, and this is indeed a world
record for solid-state track detectors. Freshly induced as
well as fossile tracks are still stable at a temperature of
$1300\,^{\circ}C$ which is only slightly lower than the fusion temperature,
$1500\,^{\circ}C$, of Oklo quartz. Very pure varieties of synthetic
quartz grown by the fusion zone technique resists annealing
of radiation damage effects only till $400\,^{\circ}C$. Researchers had
to postulate that much higher stability of Oklo quartz is
related to peculiar impurities that act as stabilizing agents.
This is a fine example of how man in his quest for betterhood
was loyal to purity and missed something, its quite natural as
ideas are pure and contain deniel of plurality.

Another interesting study is by A.I. Shlyakhter [10], who developed direct test of the constancy of fundamental nuclear constants from measured isotopic composition of Sm and E in samples from Oklo reactor core. From fission yields of Sm and Eu isotopes which had got irradiated by an independently determined fluence of reactors he calculated their capture cross-section values some 1.8 billion years ago. These isotopes were chosen because of there low lying near thermal resonances which would shift if strong interaction constant, g_S, or weak interaction constant, g_W, or Coulumb coupling constant, α, were to change with geological time, thus shifting capture cross-section values. From calculated value of capture cross-section considering error involved in their determination he showed these constants are constants within limits given below.

$$1/g_S \quad dg_S/dt \quad (yr^{-1}) \quad : \quad 5 \times 10^{-19}$$
$$1/\alpha \quad d\alpha/dt \quad (yr^{-1}) \quad : \quad 10^{-17}$$
$$1/g_W \quad dg_W/dt \quad (yr^{-1}) \quad : \quad 2 \times 10^{-12}$$

American's have shown their interest in yet another direction [11, 12, 13,14]. In their assessment, Oklo phenomeno is an excellent example of confinement of fission products, more popularly called radioactive waste these days, which had not migrated out of reaction site for around a period of 1.8 billion years. Except fissiogenic rare-gases practically everything has remained confined in Oklo reactors for 1.8 billion years. This they think has a direct bearing on the pressing problem of long term disposal of man-made radioactive waste. Oklo for

them testifies safe storage of radioactive waste in natural geological setting is feasible. Study of geochemical features of matrix in which fission products remained intact in Oklo could provide insight so that ways can be explored in which such features can be inbedded in design of nuclear waste storage facilities to minimise hazard.

As far as reactor physics aspects are concerned one extensive review [15] and six reports [16, 17, 18, 19, 20, 21] have been published in above mentioned two international meetings. These papers have dealt with criticality conditions and problem of control and sustainance of reaction. Major inclination in them have been favourable to the hypothesis of sustainance through burning up of initially present poisons. Our work banks on the hypothesis of reaction sustainance through continued mineralisation. We have undertaken to examine consequences and ground of this hypothesis.

In first chapter review of geological, geochemical and physical features of natural reactors is done. Also described is the formalism adopted to represent physical features of reactors. In second chapter is developed neutronic model for reactors. Conditions under which Oklo reactors would have become critical are discussed. In third chapter isotope burnup/buildup model is developed to study sustenance of reaction. Quantity of additional accretion of uranium is computed for sustenance of reaction till desired fluence. In chapter four is discussed general possibility of occurance of chain reaction in nature. Also discussed is conclusion and limitation of this study.

Chapter - I

GEOLOGICAL, SEDIMENTOLOGICAL AND PHYSICAL FEATURES OF OKLO REACTORS

1.1 On possibility of nuclear reaction in uranium ore:

In 1956 Kuroda[2,3] for the first time visualised possibility of natural formation of critical assembly in mines of uranium. Analysing Johanngeogenstadt pitchblende he computed ε, p, η and f as a function of geological time and established that for water to uranium ratio greater than half uranium mine would be supercritical before 2100 million years ago. Through his study he thought he had explained why ages of large scale uranium deposits never exceed 2100 million years as any such deposit at that time would destroy itself.

Discovery of occurance of large scale fission in Oklo uranium mine confirmed his visualisation but falsified his conclusion, as the fossile of reactor site is still intact and there is no evidence of destruction. And here precisely lies the mystery that how was nuclear reaction at Oklo sustained and controlled once criticality was achieved. As to how criticality was possible is in rough terms evident from Fig. 0, which gives content of uranium-235 in natural uranium as a function of geological time. Relative content of uranium-235 was higher in the past as decay rate of uranium-235 is higher than that of uranium-238. 1465 million years ago uranium-235 enrichment was 2.3%. This would mean Tarapur Reactor could be fueled by natural uranium at that time. Also any assemblage of concentrated uranium could go critical with natural water around or before that time.
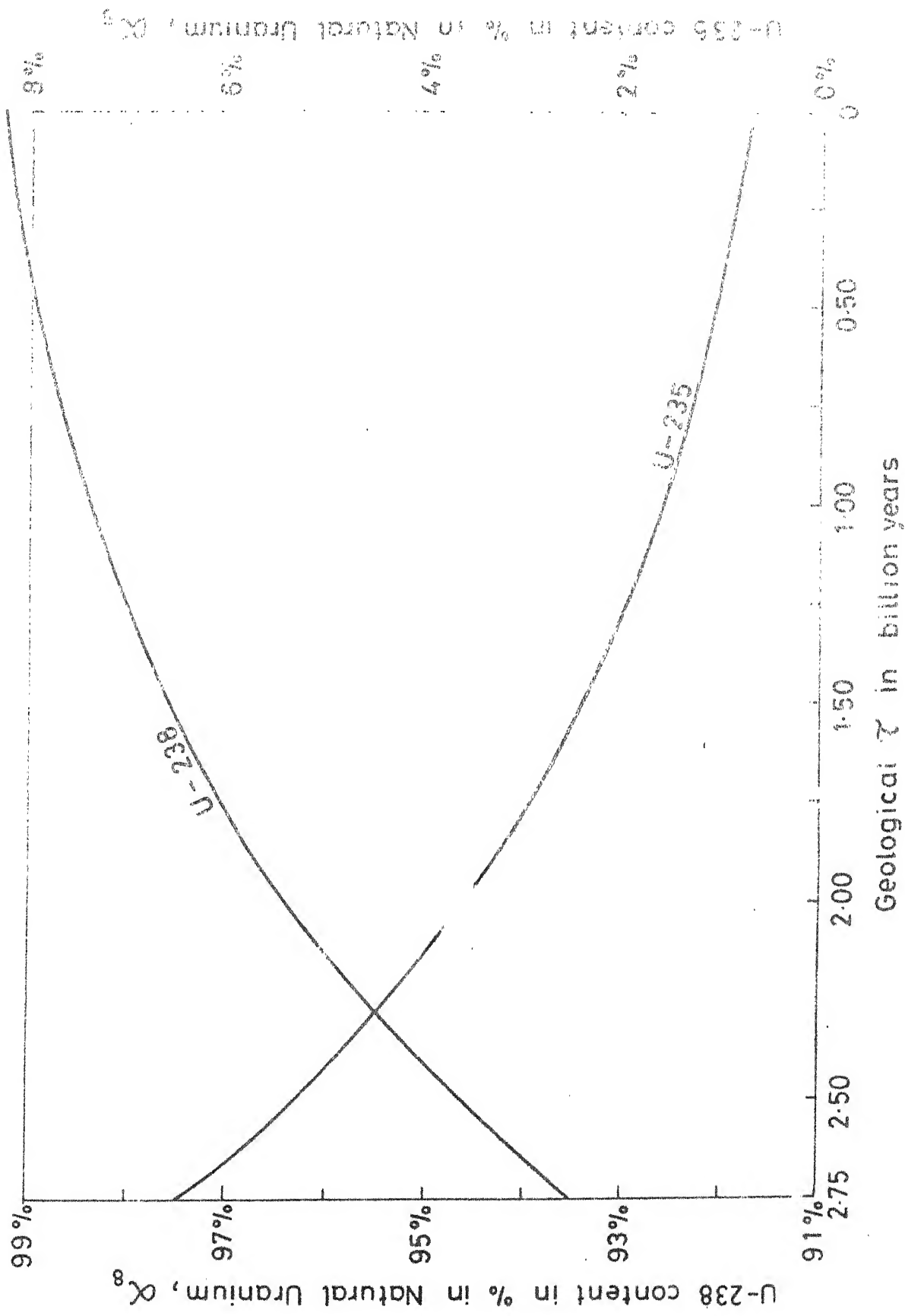
Fig. 0 Variation of Uranium enrichment with Geological Time

To go into details of working of enearthed natural reactors we will have to trace origin of Oklo deposits and subsequent evolution leading to their working. As the physical history of reactors can not be separated from geological history it would require review of geological and geochemical aspects which were central to unfolding of this phenomena.

1.2 Geological and geochemical features :

In Table I results of several studies regarding age of uranium deposition have been compiled. Average of these esti-mations comes to 1905 ± 151 million years. The reactors must have worked during or after this time.

Oklo mineralisation geologically was an terrain phenomena a thorium content in uraninite crystal is very low. This is only possible if deposition took place from very low temperature (around 70°C was surface temperature at that time) aqueous solution because thorium is insoluble in low temperature aqueous solution [22]. Also preservation of fossil algae in the host Francevelle series is indicative of that [23].

About 2000 million years ago land mass over earth looked quite different. South America was close to Africa and Oklo was situated at the ridge of two evolving continents. Fig. 1 shows position of Oklo in the global map at the time of ore deposition.

TABLE 1

| Age of Mineralisation (τ) in Million years	Method of study
1800-2000	K/Ar isotopic study [42]
1750	U/Pb isotopic study [43]
1800-2050	U/Pb isotopic study [44]
2000	Nd,Sm,Ru,Th isotopic [44] abundance study
1600-2100	Parameter fit model [41] of Nd isotopic abundance
1800-2100	For different Thermal[45] fluences published Nd^{143} isotopic study
2000 ± 100	From Uranium fission [46] data
2050 ± 30	U/Pb study [47]
1730	For motion age of [48] quartz crystal from fission track analysis
1700-1800	K/Ar and Rb/Sr isotopic [49] analysis
1800	K/Ar and Rb/Sr isotopic [42] analysis
1930 1900	Th/U/Pb isotopic analysis[51]
1980 ± 500	Nd isotopic study [51]
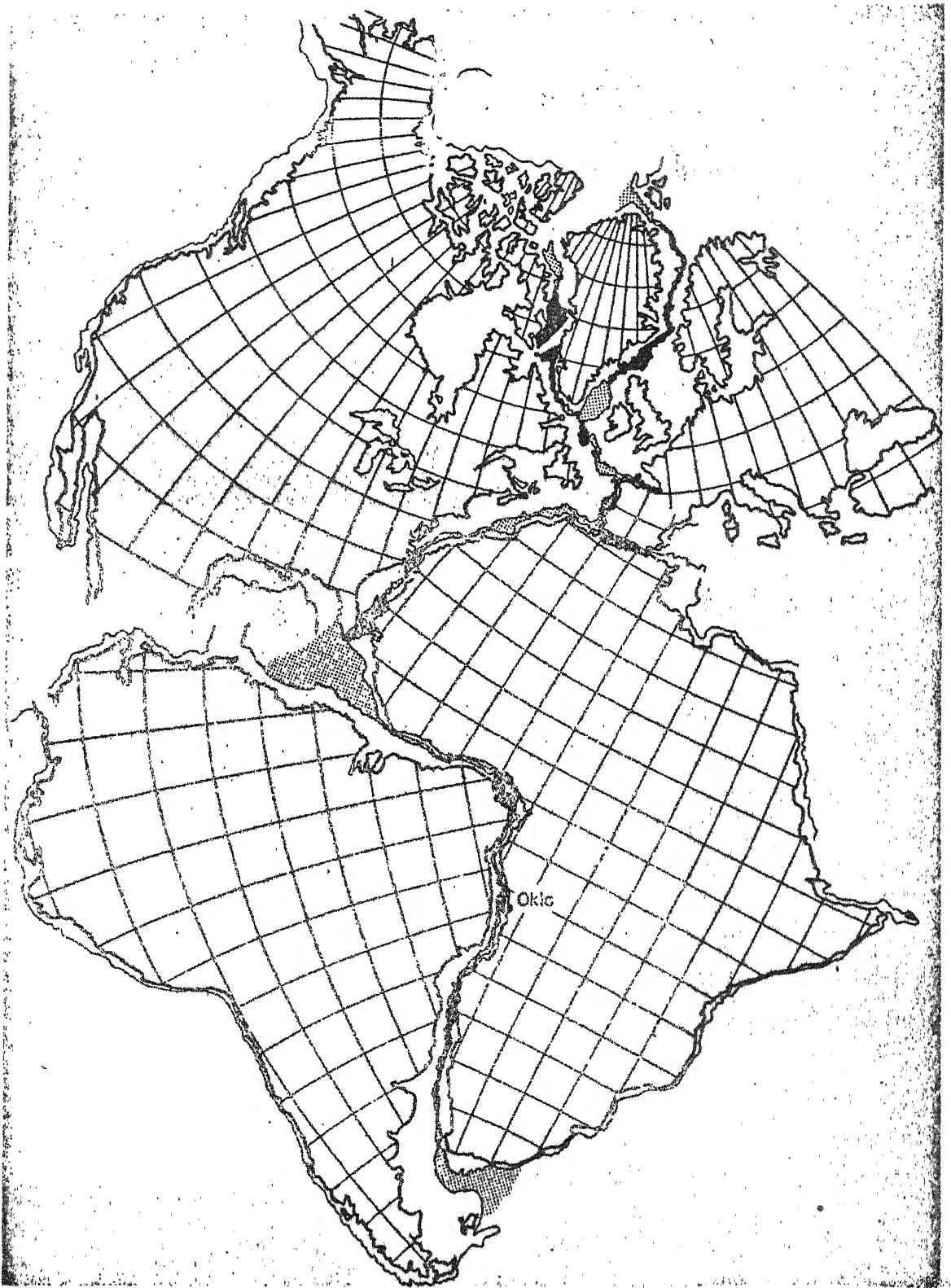
Fig. 1. Location of Oklo in the Republic of Gabon, Africa. The continents are shown in the positions which they occupied 2×10^9 years ago, wh..n the Oklo reactor was operating.

African continent is made up of four consolidated and granitized Cratons. The West African, Congolese, Kalahari and Nilotic Cratons are represented as region A, B, C and D respectively in Fig. 2(a). With these Cratons are associated internal "intracratonic" basins with shallow detritic sedimentations and intercratonic zones with their own deep sedimentations. Intracratonic zones are located at the periphery of the cratons on the fold axes and are called 'mobile zones'. Gabon lies in the north-west part of the Congolese Craton. Francevelle basin which is in south-east of Gabon, and in which lies Oklo is one of the intracratonic basins of the Congolese Craton and is marked 22 in Fig. 2(b).

The present structure of Africa only got established by the end of precambrian era (500-600 million years ago) [24]. Before that geological history of Africa is marked by series of brief paroxysmal events in prolonged periods of stability. Surfacial geological cycle of erosion, transport, sedimentation and folding is called Orogeny. The final paroxysmal phase is called orogeneses. Chronologically in Africa there have been four major orogeneses : Precambrian A (500-600 to 900-1200 million years ago), Precambrian B (900-1200 to 1800-2000 million years ago), Precambrian C (1800-2000 to 2500-million years ago), Precambrian D (before 2500 million years ago). The Francevelle basin can be assigned to the precamberian B orogeneses as age of deposition for it falls in that period.
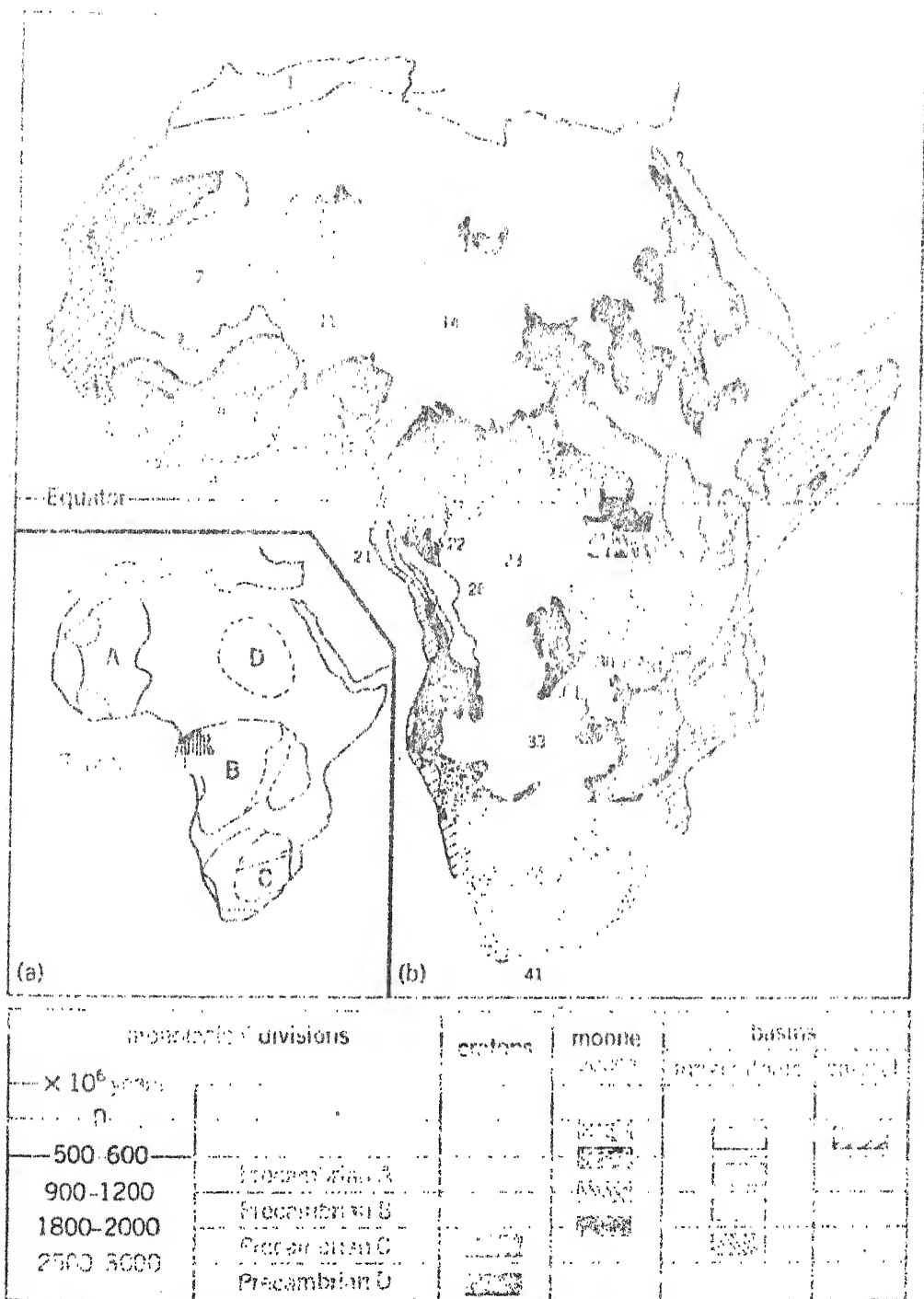
Fig. 2. Geological map of Africa. (a) Cratons. A = West African craton, B = Congolese craton, C = Kalahari craton, D = nilotic craton. (b) Principal structural units indicated by numbers. (From P. Molina and J. C. Besombes, The Oklo Phenomenon, International Atomic Energy Agency, Vienna, 1975)

Through an independent study also one can extablish that uranium ore formation in sediments could not be older than precambrian B orogeneses. Before 2200 to 2000 million years ago earth's atmosphere had little oxygen. Because of lack of oxygen, surfacial uranium in volcanic ashes and Basaltic rocks was highly immobile as uranium existed in U^{4+} chemical state, in which it is insoluble in water, and only in its oxidised state U^{6+} it is soluble in water. From chemical immobility of uranium in U^{4+} state we can infer that uranium existed in very low concentration (about $10^{-4}\%$ by weight) in volcanic ashes and older Basaltic rocks. Around 2200 to 2000 million years ago rise of new form of becterial life called blue-green algee, which was the first organism capable of photosynthesis, liberated oxygen in large quantities. Thus making earth's atmosphere oxygenic also increasing content of dessolved oxygen in water. This was not only important for future evolution of life but also played irreplaceble role in oxidising uranium to U^{6+} state enhanced by the process of surface leaching, by the process of surface weathering because of torrential rains, and by rock erossion because of winds. This only made possible mobility of uranium as Uranyl ion, UO_2^{2+}, along with metals having similar geochemical behavior. With its host, water, it move around surface of earth along rivers, infiltrating sediments till it got reduced in geological traps back to U^{4+} state [9]. For uranium geological traps in sediments were locations with high reduction potential where uranyl ion got detained from moving water. If

conditions had been such that water flowed through traps for long time uranium concentration in them would increase to make those sediments uranium ores of future. These series of phenomena not only explain formation of uranium minerals but also explain why there is no large scale known surfacial deposit on earth older than 2100 million years.

Existance of concentrated localised carbonacious matter in sedements could have been ideal reduction medium for uranium. And fossilised life colony or organic ooz in newly formed sediments cross-cutting waterbed probably provided such condition for formation of uranium ore in Francevelle basin. Four such large locations illustrated in Fig. 3. have been unearthed in Francevelle basin, those of Boyindzi, Mounana, Oklo and Okelo-bonde. All of them are situated in a same flexural zone effected by sliding. First two, at the contact of basement are subject to break deformation, while the other two are implicated in extensive flexible structure resulting from same sliding tectonics. Oklo uranium deposit is situated 1500 m south of Mounana deposit. Concentration of uranium is these large zones would'nt have gone more than 0.4% to 0.6% by weight in primary mineralisation [25]. Though there would have been little variation in degree of mineralisation due to locally variable sedimentological conditions. Geological studies indicate that primary mineralisation stopped because of continued sadimentation as mineralisation zones reached depths inaccessible for uranyl rich water to penetrate it [26].

Fig. 3. Zone of primary Uranium mineralisation in Francevelle Basin

Upper figure in Fig. 4 maps zone of primary minerali-
sation in Oklo projected on horizontal plane as seen vertically
where as lower figure is a sectional view along vertical planes
marked A-B, in upper figure. In Francevelle basin there has
been two superposed sandstone-pelite cycles. In the fore deep
basin uranium is localised in a 4 to 10 m thick layer in upper
part of basal sandstone. Maximum size of mineralisation zone
is 900 to 600 m. Figure 4 illustrates situation of deposit
today. Actually uranium mineral went through long history of
tectonics and further geochemical rearrangements before aquiring
this shape. One unique feature of Oklo as distinct from other
three deposits in Francevelle is that it is close to basal rise
which conditioned tectonics and influenced sedimentations. Fig.5
illustrates how primary flat mineralisation zone because of
future tectonics underwent change. This whole process of pre-
cambrian B orogenesis must have taken millions of years before
leading to present rock shape. These tectonic movements lead
to formation of extensive shear troughs in Oklo mineralisation.
Whereas in Boyindzi and Maunana sedimentary rock got fractured
and in Okelebonde little sliding occured. The formation of
shear troughs played unique role in the formation of local
tectono-sedimentary traps in primary mineralisation zone which
lead to reconcentration of uranium in those areas after mineral
sediments reached deep burial depth. Pressure and temperature
conditions at that depth coupled with paroxysmal tectonic activity
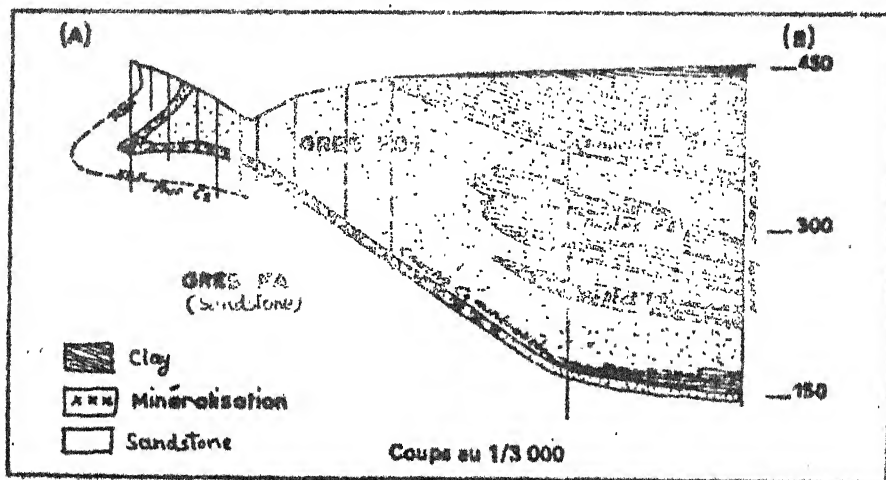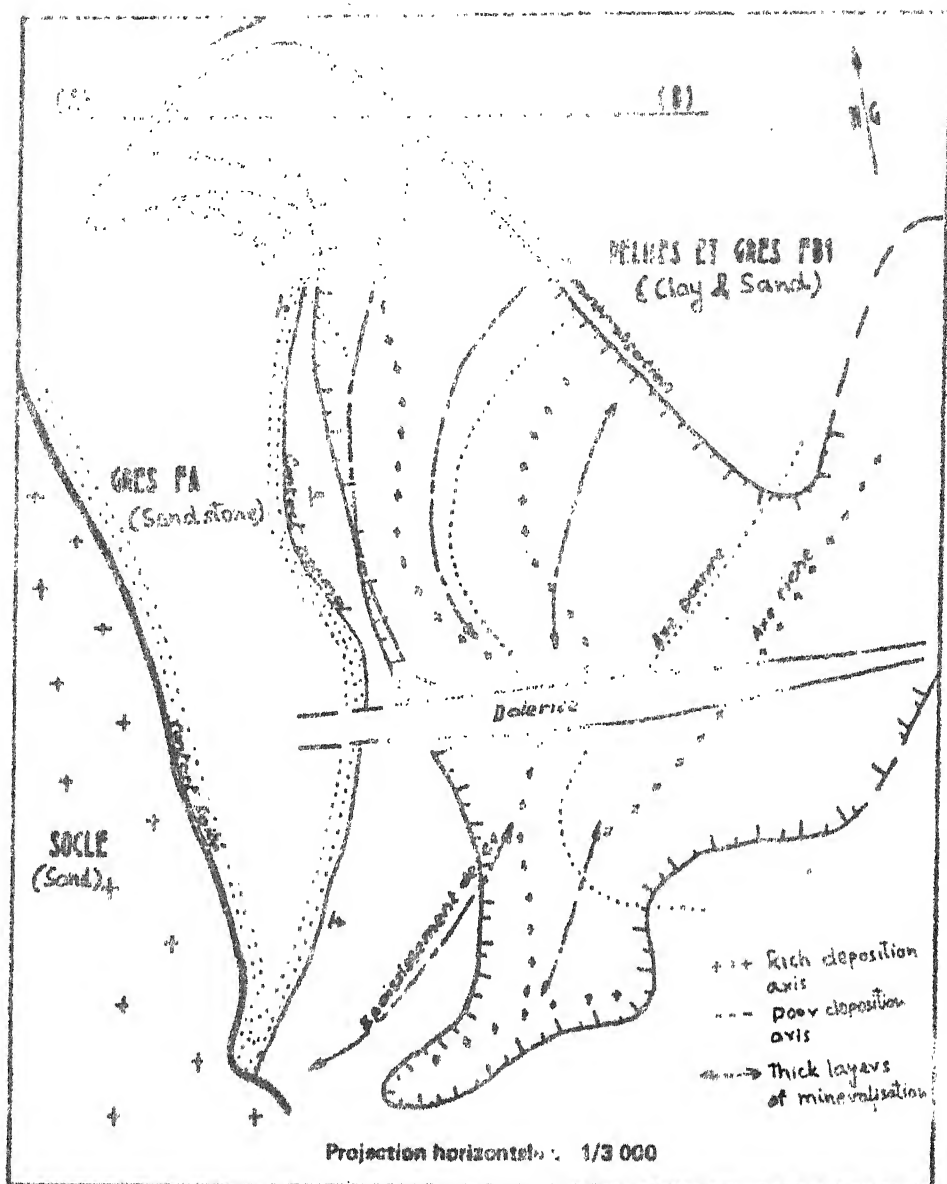lead to total desilicification of sandstone by oxidising currents

Fig. 4. Mineralisation zone of Oklo
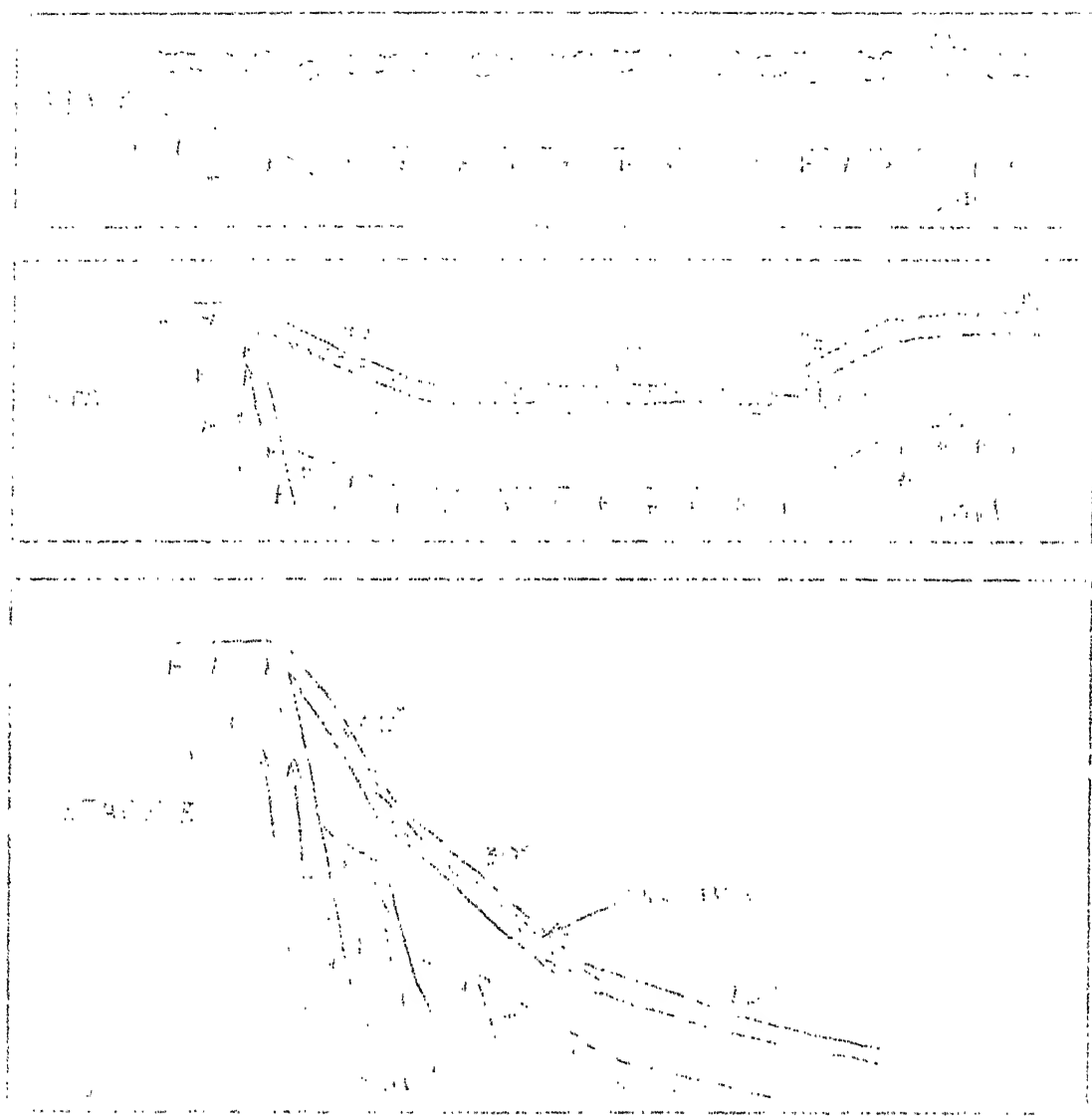
Fig. 5: folding of mineralisation zone by tectonic activity

in shear troughs and uranium got concentrated in extension zones forming anamolous uranium rich lodes within large normal mineral zone. When this process of concentration of uranium was going on at some point eventually criticality was attained. This silicon diagenesis lead to compaction and settling of earth to the degree in which we find Oklo ore today. Evidence for this diagenesis is substantiated by patrographic studies [27] which indicate two distinct type of uranium ore in Oklo with distinct modes of formation. In normal ore uranium appears mostly as micro-veinlets of pitchblende, with corresponding mean uranium concentration about 0.5%, embedded in a matrix of clay mineral rich in organic matter. In deposition of this type of ore predominent role was played by organic material. In the anamolous uranium rich zones sandstone is predominantly chlorite rather than clay and uranium appears as a cluster of tiny ($\leqslant$ 100 µm) and well crystalised uraninite grains and uranium shows about 50 fold increase in concentration over 0.5% value in normal ore. Certain spots even have uranium concentration upto 60%. Migration of oxidising solution could only be the main source of reconstruction. And this would need moving out of silica to make place for uranium.

Six sites of these compact accumulation of very rich uranium ore has been identified in Oklo. All of them virtually lie in a single block flattened in the direction of stratum. Average thickness of these lodes is about one meter and their length about 10 to 20 meters. Figs. 6 shows horizontal

Ra. >44.000 c/s.

Ra. 10.000 to 22.000 c/s.

Ra. 22.000 to 44.000 c/s.

Echelle 40 m
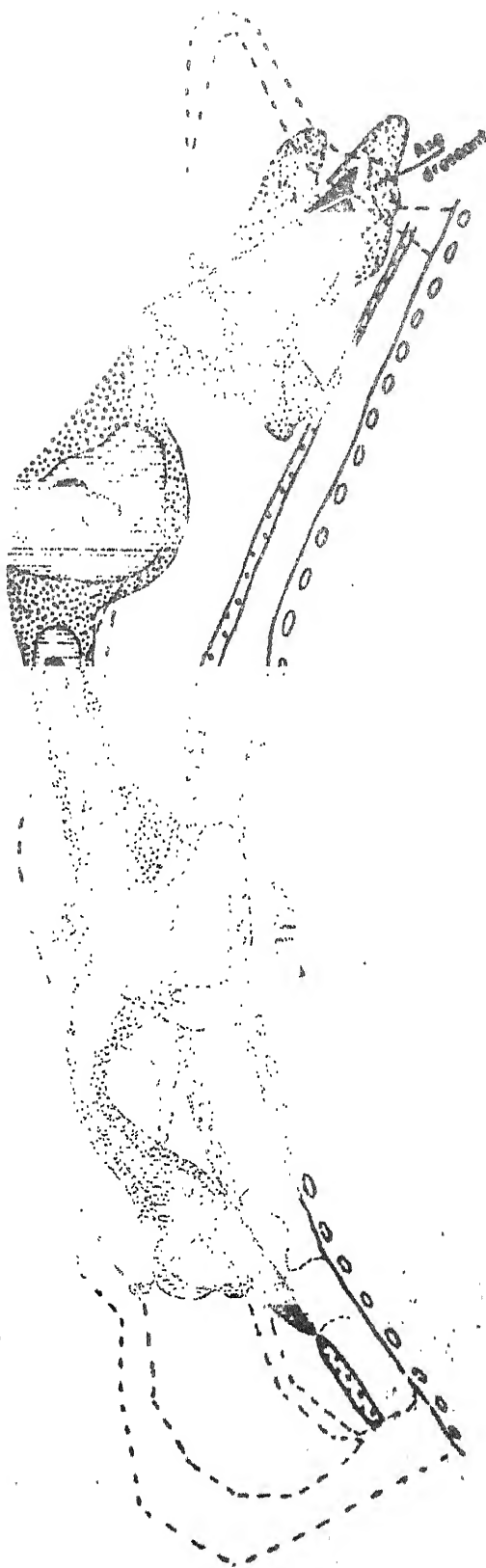
Ra > 44,000 c/s

Ra 22,000 to 44,000 c/s

Ra 16,000 to 22,000 c/s
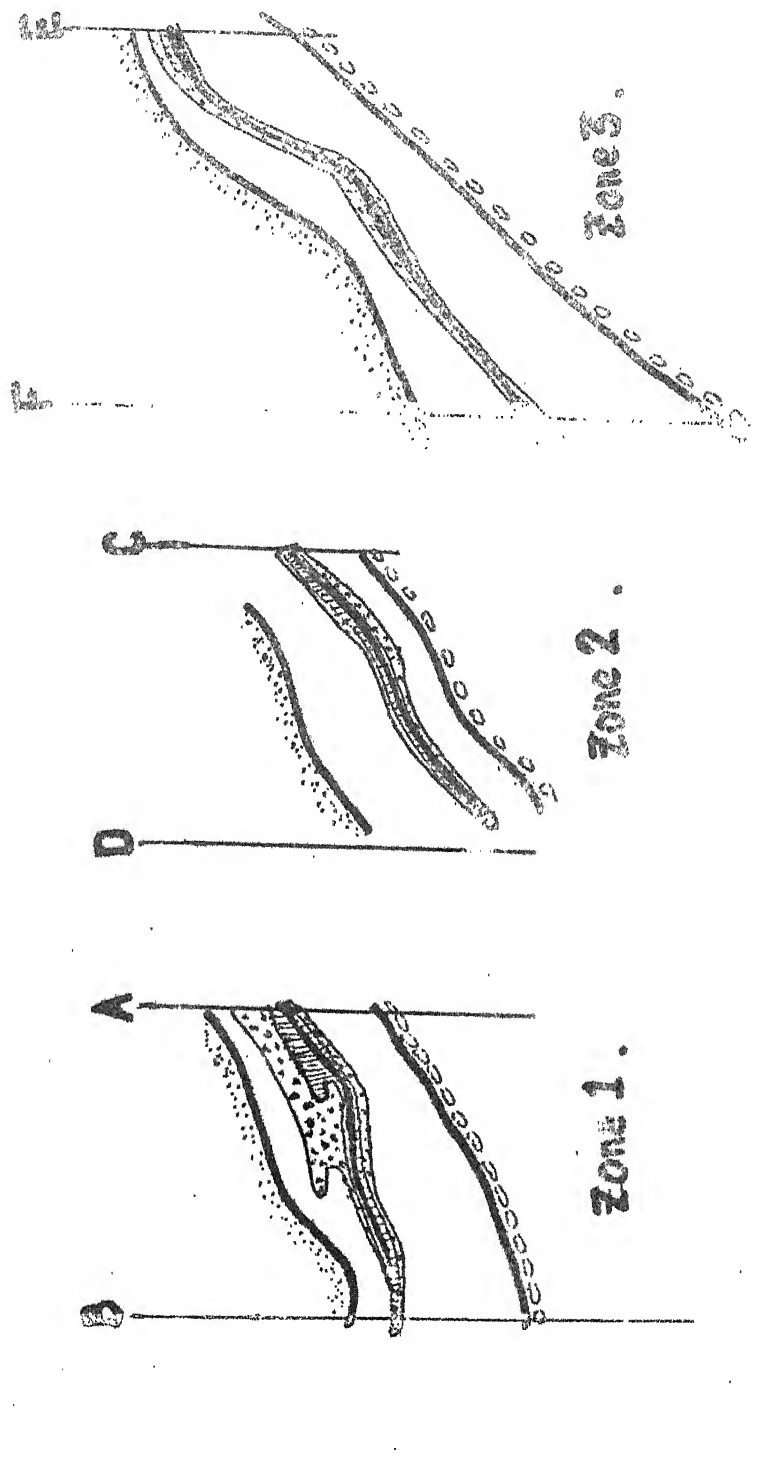
Conglomerate

Clay

Zone 1.

Zone 2.

Zone 5.

Scale: 5m

Fig.7: Sectional view of reaction zones along vertical planes marked A–B, C–D, E–F in Fig.6.

projection of these zones maped according to the radioactivity
counts in radiation counter, indicating uranium concentration
today. Fig. 7 is a sectional view of these compact zones along
vertical plane A-B, C-D and E-F. Total surface area of these
zones is about 1.5% of surface area of primary mineralisation.
Average uranium concentration of these zones is greater than 10% by
weight today. It was only in these zones that marked depletion
of uranium-235 is found. This would imply that nuclear reaction
took place only in them. Certain tectonic episode coupled with
continued increase in buriel depth had triggered their formation
within primary mineralisation. This was made possible because
of uniqueness of rock structure of Oklo deposits. Fig. 8 illus-
trates fold structure of Oklo which streches 500 meter in North-
South direction and reaction zones are shown at base of these
folds lying in the extension zone of Dome slightly away from
the main slippage. Rise of this Dome was central for formation
of uranium rich lodes and attainment of criticality in them.
When reaction began it resulted in local thermal metamorphism.
Thermal syphoning progressively expelled water and organic
matter from core of the reaction zones. In this process coarsely
crystalline uraninite was formed and eventually burning up of
uranium brought reaction to end. After finishing up of criticality
settlement of earth took place once again in which carbon played
dominant role that is why this phase is called carbonate diagenesis.

Geological history since primary mineralisation has been
illustrated in Fig. 9. Depth which mineral sediment reached has

FRACTURE

REACTORS

PARTIALY EXPLOITED
REACTOR ZONE 1 & 2.
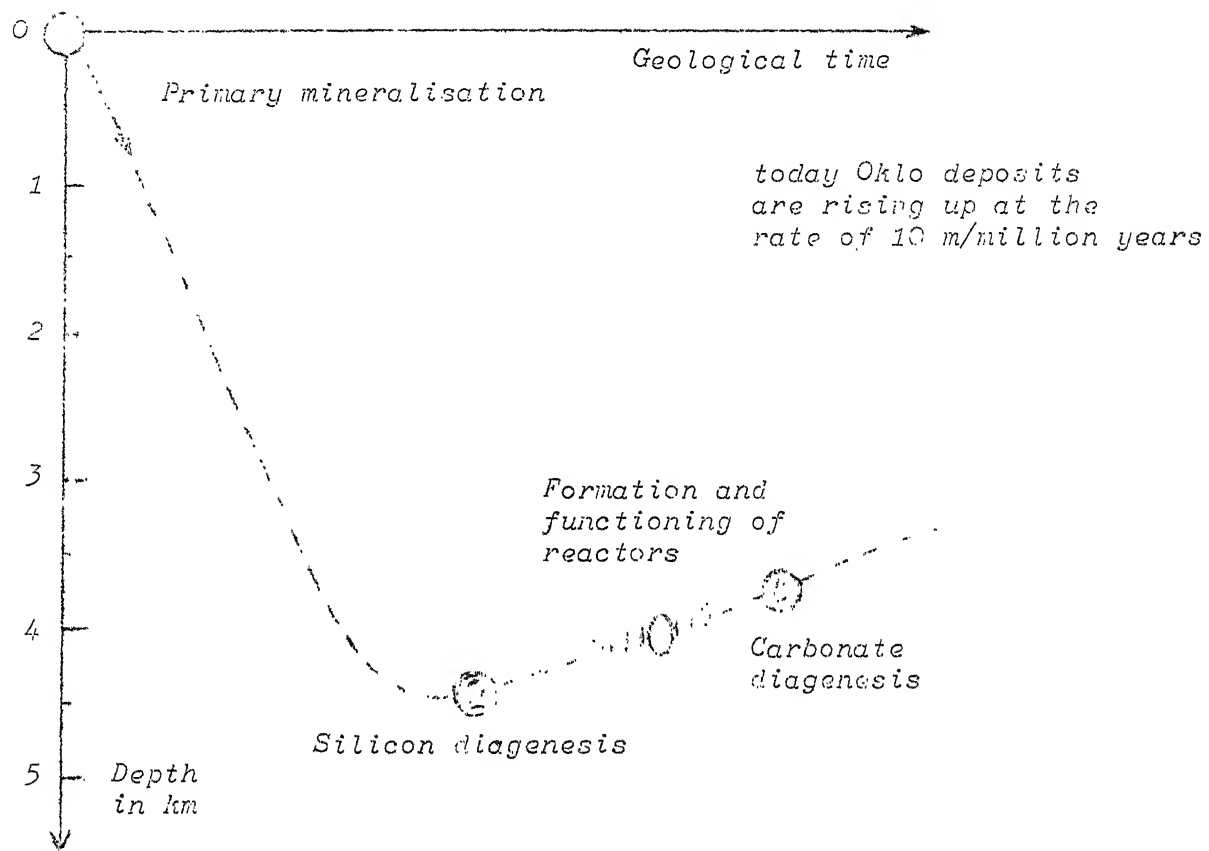
Fig. 9: Geological history since primary mineralisation.

been obtained from working out of tectonic history and also by fission track studies [9]. It was in the later part of desilicanisation process that uranium got concentrated enough to start chain reaction. At that stage reactor was buried 4.8 kms deep. Generalised variation of Lithostatic and hydrostatic pressure with depth has been plotted in Fig. 10. Dotted line indicates pressure condition at the finish of desilicanisation process. In Fig. 11 is plotted pressure temperature geothermal gradient at Oklo. Line 3 is an isochore of the fluid in rock contemperary to the formation of primary vien of quartz at the begining of desilica diagenesis. Line 2 is an isochore of fluid contemperary to the formation of final quartz at the finish of nuclear reaction. Line 1 is isochore contemperary to the carbonate diagenesis after reaction site cooled down geologically. From these curves one can infer that temperature of fluid at the time of establishment of criticality must be between 180°C to 230°C. If surface temperature in precambrian times was more than 50°C as one study indicates surface temperature to be 70°C to 80°C than temperature of sediments would be 210°C to 250°C. Cross-sections used for studying neutronics have to be modified for this temperature.

1.3 Uranium content in reaction zones :

In Table 2 are compilled content of uranium in reactor zones as well as amount of uranium-235 depletion observed [35]in reactor sites today. From these figures amount of uranium-235
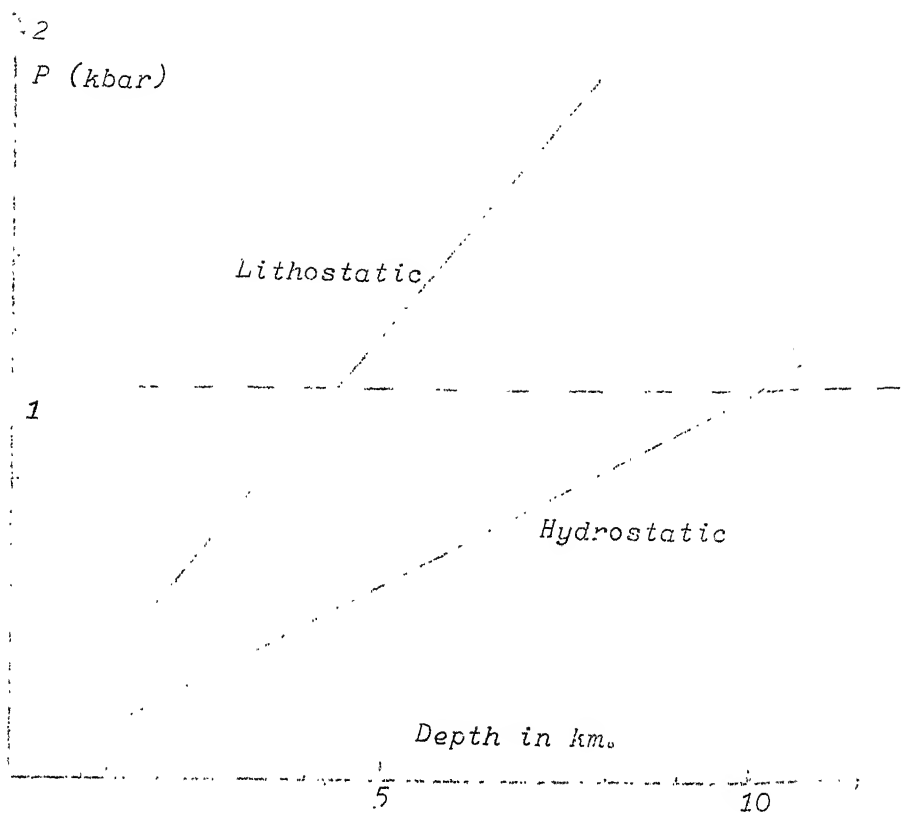
Fig. 10   Lithostatic and Hydrostatic pressure variation with depth
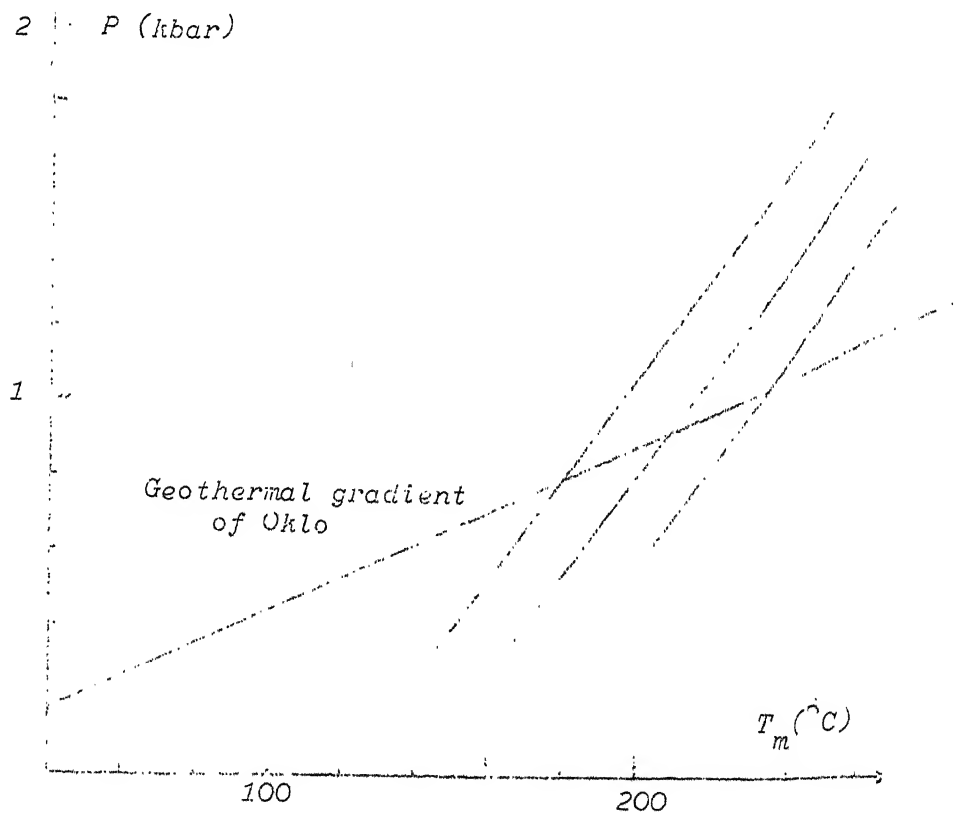
Fig.1 :   Geothermal gradient of Oklo

TABLE II

Reaction zone	Amount of Uranium-235 missing today	Amount of Uranium-235 burnt (If reactor worked 1.9 billion years ago)	Total quantity of Uranium involved in Reaction	Total amount of material in the core (Ass: 30% uranium)	Volume of core	Geometric buckling Assumed geometry
1	200 Kg	1.27 tonn	230 tonn to 240 tonn	561 tonn to 585.4 tonn	137 m³ to 142 m³	$10.01 \ m^{-2}$
2	200 Kg	1.27 tonn	200 tonn	488 tonn	119 m³	$10.04 \ m^{-2}$
3-4	90 Kg to 100 Kg	0.64 tonn	180 tonn to 190 tonn	440 tonn to 465 tonn	107 m³ to 113 m³	$10.06 \ m^{-2}$
5-6	100 Kg to 110 Kg	0.7 tonn	170 tonn to 180 tonn	416 tonn to 440 tonn	101 m³ to 107 m³	$10.07 \ m^{-2}$

burnt if reaction took place 1900 million years ago have been
deduced. Also total amount and volume of materials in these
zones have been tabulated with the assumation that 30% of
material in reactor zones was uranium. Idealising reactor zone
shapes by one meter thick slab with square surface, geometric
Buckling has been calculated. For all the reactor zones it is
slightly more than 10 m^{-2}. For reactor systems 3-4 and 5-6
the surface area will be much more than idealised geometry area.
This would imply geometric buckling would be higher than calculated.
If reaction had began originally in smaller volume and grown to
the volume observed today than again geometric buckling would
be higher first and then will reduce to 10 m^{-2} by closing of
reaction.

Measurement of content of uranium, U%, and partial
density, d_u, of uranium were carried out for various samples
from ore site. Partial density is defined as weight of uranium
per unit volume of sedementary rock. Result of the survey[28] is
plotted in Fig. 13. For analytic purpose best fit curve can be
idealised by following equation:

$$\frac{1}{d_u} = 0.52 - 0.48 \times U\% /100 \qquad (1)$$

From above equation one can find out number density of
uranium for any value of content of uranium ranging between 10%
to 50% by weight.
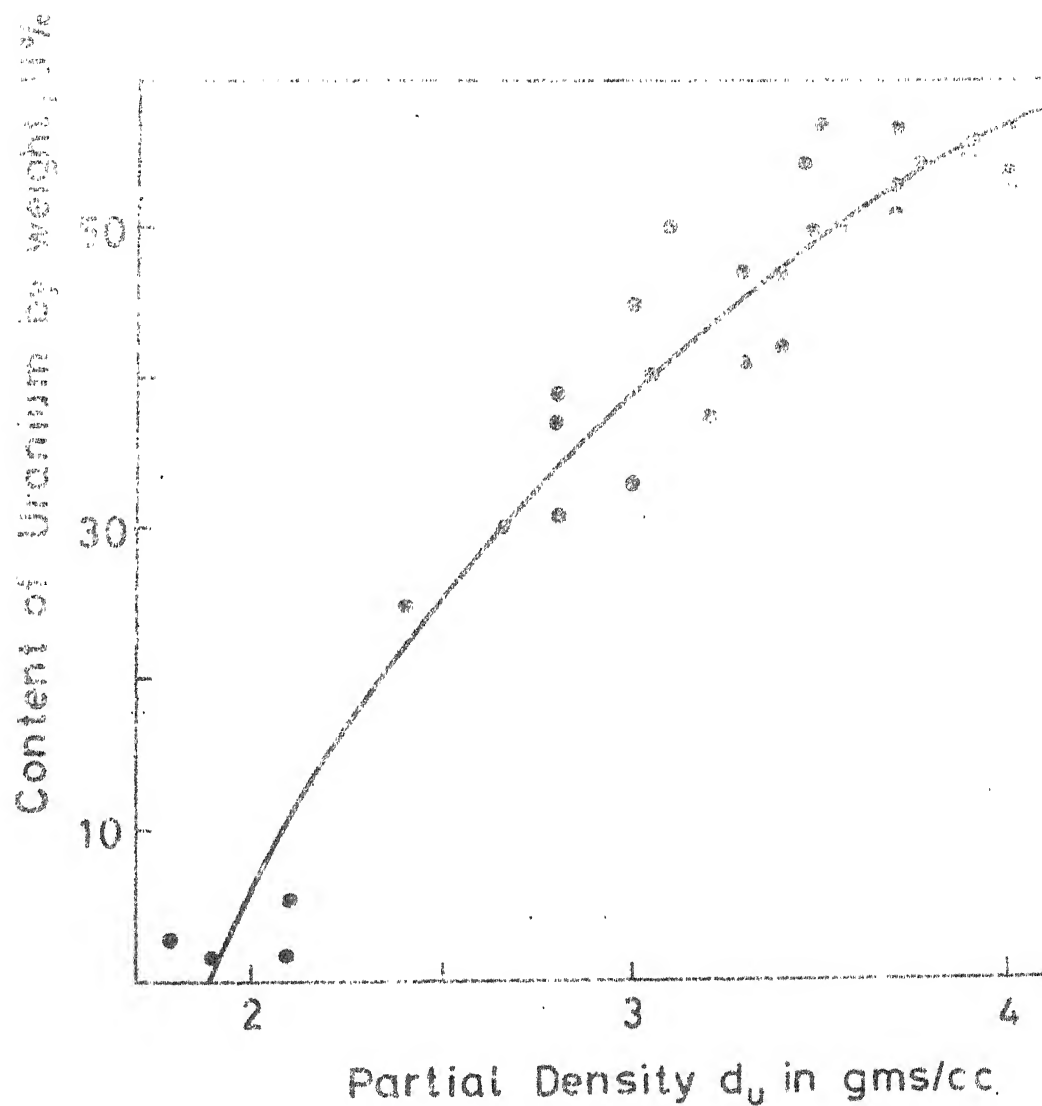
Fig. 13   Relation of partial density of Uranium and content of Uranium by weight in Oklo rock

1.4 Content of rest of material (gangue) in reaction zones :

From the same survey content of rest of material in reaction site as a function of uranium content in rock and geological time is infered as [16]:

$$X = \frac{100 - 1.235 \times U\%}{1.324 \times U\%} \times \exp(-0.175\tau) \qquad (2)$$

where X is defined as weight of gangue per unit weight of
UO$_2$ in same volume,

and τ is geological time in billion years.

Exponential term comes from taking into account of transformation of uranium into lead by natural decay. Other terms come because gangue does not include water which is chemically bound with mineral also a factor comes because weight of UO$_2$ is different from weight of uranium.

1.5 Content of different constituents of gangue :

Silicon and Aluminium are major constituents of Oklo rock as clay and sand were host sediments where mineralisation took place. Magnesium, Iron, Manganese, Potassium and Calcium are easily traped by organic rich lences along with uranium from low temperature aqueous solution. These are the elements which constitute most of the gangue though there are thirty odd elements found in rock. Average of measurement of relative content of different elements in reactor zone 2 are as follows:

SiO_2 42% ; Al_2O_3 28% ; FeO 10.5% ; Fe_2O_3 4.5%

MgO 7.5% ; K_2O 2.5% ; CaO 2.5% ; Na_2O 0.6%

TiO_2 0.7% ; MnO 0.5% ; BaO 0.2% ; Others 0.5%

From neutronics point of view content of important trace elements was found out to be following

Vanadium 200-800 ppm ; Cr, Ni, Ca 30-100 ppm

Boron 20-250 ppm ; Sm 10-30 ppm

Gd 7-20 ppm ; Li 100 ppm

Hf 20 ppm

For simplicity and to develop further analytic scheme constituents of gangue can be divided by inference from actual survey as given below [16]:

$$SiO_2 \; : \; 44.5 - 0.05 \times U\%$$

$$Al_2O_3 \; : \; 30.5 - 0.05 \times U\%$$

$$MgO \; : \; 11.7 - 0.08 \times U\% \qquad\qquad (3)$$

$$FeO \; : \; 10.0 + 0.15 \times U\%$$

$$K_2O \; : \; 2.5$$

$$MnO \; : \; 0.0 + 0.01 \times U\%$$

These relation have been derived by research team headed by Neudet [16] from actual measurement of content of different element oxides in rock samples with different uranium concentration. Also these relations have been checked by study of

geochemistry of desilicanisation process. In above given set of equations contents of SiO_2, Al_2O_3 and MgO are a bit over-estimated to normalise but because of their low scattering c cross-sections these overestimations account for other elements neglected. MnO is also overestimated but because of its high capture cross-section it takes care of other elements neglected. FeO estimate is low as Fe_2O_3 and FeO have been bracketed and their combined scattering can be better estimated by taking slightly low FeO content.

Number densities of constituents in gangue can now be written in simple consistant analytic form as function of content of uranium and geological time.

The trace element in gangue which have high neutron capture cross-section have to be separately taken into account. Macroscopic absorption cross-section of all of them can be represented as equivalent to some quantity of B^{10}. Isotopic geochemical studies indicate their estimated content in reaction zone could have been equivalent to 50 to 250 B^{10} atoms per million atoms of uranium [16].

1.6   *Water content in reaction zone :*

There is about 9% to 11% water by weight in gangue which is chemically bound with gangue. There is also possibility of existence of water in the pores of rock. In fact hypothesising existance of water in rock pores when sediments were buried deep is vary essential to the thesis of desilicanisation and decorbonate

diagenesis. During primary mineralisation, porosity planes, where mineralisation took place, must have been saturated with water. As the buriel depth of sediments increased there was possibility of water boiling out as precambrian surface temperature was about $70^{\circ}C$. In that case whatever water remained in rock pores could have at deep buriel depths moved into shear troughs formed by tectonic activities and it was there that reconcentration of uranium took place. It seems quite acceptable that rock pores in reaction zones must have been filled with water. Estimation of volume of rock pores is difficult. But generally 6 to 12% volume of rocks is microfractures for old sedimentary rocks. Two kind of water in Oklo rock can be formalised in following way to express content of water.

$$Y \equiv \frac{\text{weight of water/volume}}{\text{weight of } UO_2/\text{volume}} = Xm' + m$$

where m' is acontent of water chemically bound to gangue per unit weight of gangue. Xm' gives weight of chemically bound water per unit weight of UO_2 for unit volume. m is weight of water in pores per unit weight of UO_2 in unit volume.

Weight of loose water in 100 gms of rock is

$$= 1.34454 \times U\% \times m$$

If d_w was density of water in those condition and d_r was density of rock then volume percentage of loose water is

$$V\% = 1.345 \frac{d_r \times m \times U\%}{d_w}$$

This has to be always less than volume fraction of rock pores. Now expression for water content becomes

$$Y = Xm' + 0.744 \frac{d_w x V\%}{d_r x U\%} \quad \text{where } V\% \in [6\% \text{ to } 12\%]$$

Water is pores during and after desilicanisation process was subjected to pressure between 800 bars to 1000 bars. and its temperature was $150^\circ C$ to $300^\circ C$. Water in this region is supercritical and its density varies smoothly with temperature[40] as shown in Fig. (14). In the region of interest straight line approximation can be made to analytically formulate temperature dependence of loose water density.

$$d_w = -9.4 \times 10^{-4} T_m + 1.106$$

where T_m is temperature of milieu in degree centigrade. Now expression for Y becomes

$$Y = Xm' + 0.744 (-9.4 \times 10^{-4} T_m + 1.106) V\%/(U\% d_r)$$

$$(4)$$

Here we have formulation of Y which is sensitive to measurable and physically constrained parameters $V\%$, d_r, m'. m' was averaged over reaction zone 2 and was found to be close to 0.10 (10% of gangue). Density of dry Oklo rock has been estimated to be between 2 to 3 gms/cc for varied uranium content. Fig. (15a,b,c,d) shows variation of Y with $V\%$, $U\%$, m', d_r at temperature $200^\circ C$ 1.8 billion years ago. With most optimistic combination of different parameters Y could not
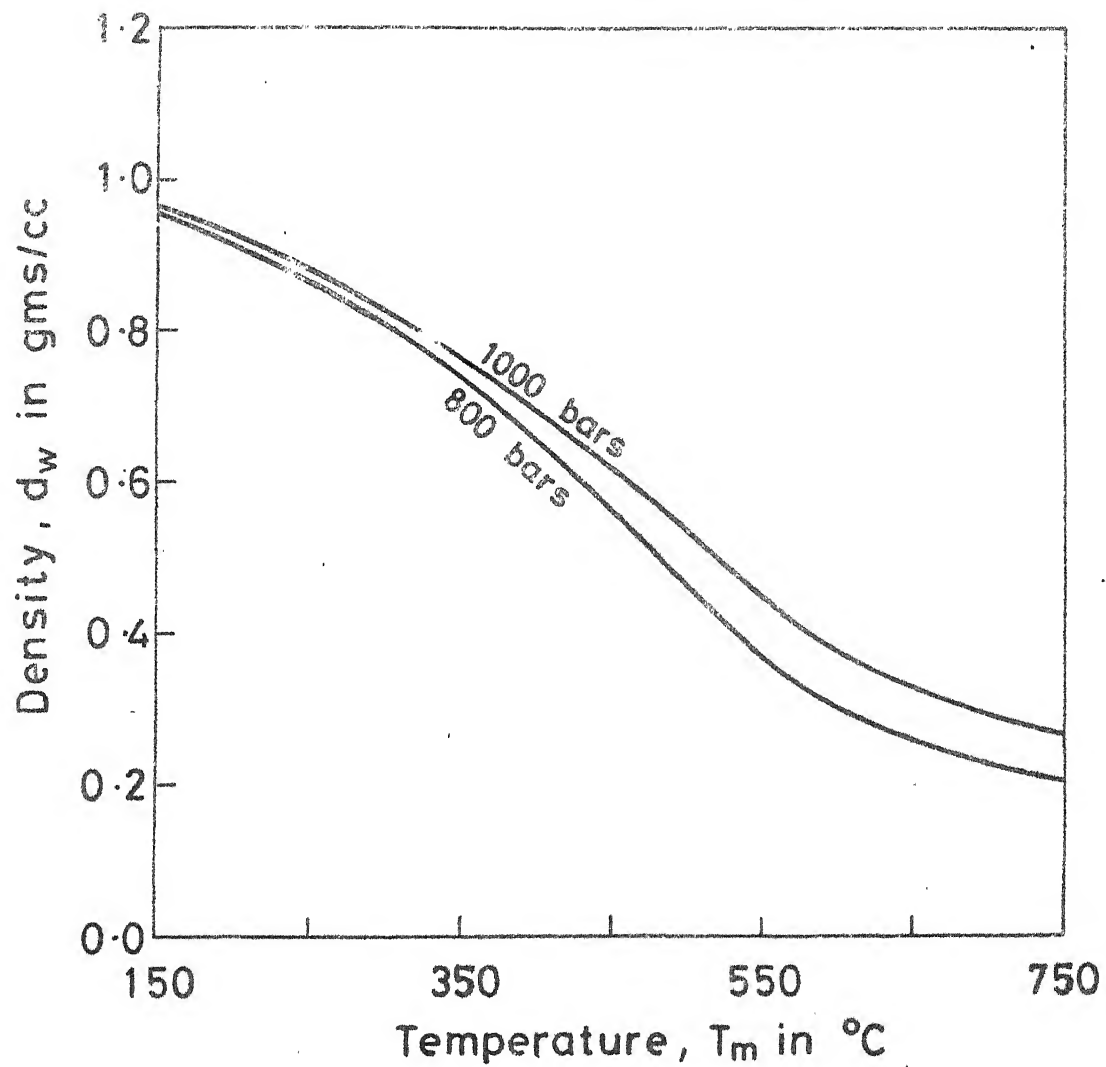
Fig.14 Density variations of supercritical water with temperature
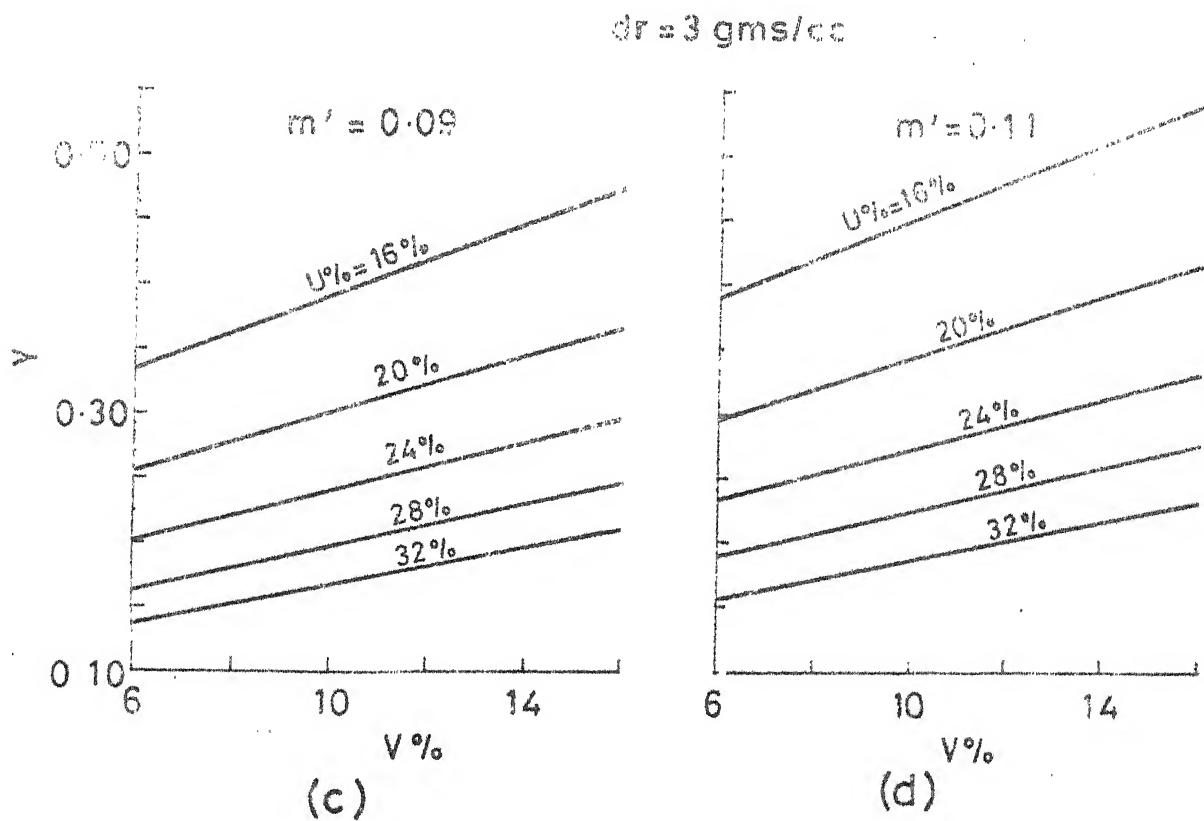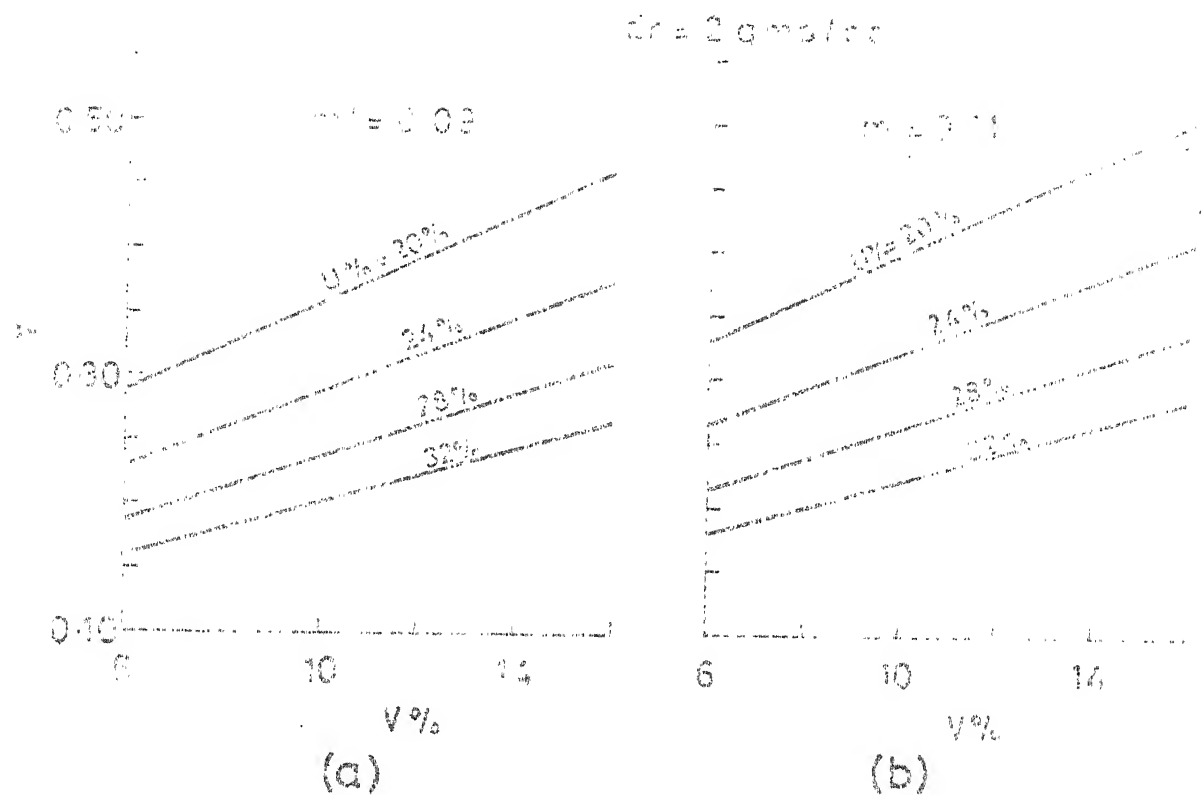
Fig.15 Content of water vs volume content of loose water

have been greater than 0.5. If we take volume fraction of loose water to be about 10% (= V%) and uranium content to be around 24% (= U%) as was in most of Oklo reactors, Y turns out to be less than 0.32 for d_r = 2 and less than 0.28 for d_r = 3. By simply averaging densities of components of rock we can estimate that density of rock is closer to 3 gms/cc and we can say with some certitude that Y could not have been greater than 0.3 in Oklo reactors. For higher uranium contents it is even less.

STUDY OF CONDITIONS OF CRITICALITY

2.1 *Reactor physics model :*

In this chapter reactor physics model for Natural reactors of Oklo has been constructed which will enable us to assess conditions under which natural assemblage of uranium could go critical. For evaluation of criticality conditions we would work with the assumption of homogenised medium disregarding spacial distribution of uranium and other materials in Reactor zones as well as whatever lumping of uranium exists in them. Also we would work with the assumption that all material in reactor zone is exposed to same neutron flux.

For any assemblage of fissile material to sustain neutron chain reaction effective neutron multiplication factor (K_{eff}) should be one or greater than one. From homogenious thermal diffusion equation and Fermi Age theory expression for K_{eff} is given by following equation [36].

$$K_{eff} = \frac{k_{inf}}{(1 + L^2 B^2)} \exp(-\tau' B^2) \qquad (5)$$

where

B^2 is geometric buckling of assemblage

L^2 is diffusion area for thermal neutrons

τ' is Age from fast to thermal energies

K_{inf} is infinite multiplication factor. It is an intrinsic

property of materials in Reactor and is given by the product of four factors.

$$K_{inf} = \eta \cdot f \cdot \varepsilon \cdot p \qquad (6)$$

Four factors are:

η is regeneration factor

f is thermal utilisation factor

ε is fast fission factor

and p is resonance escape probability.

Now we will elaborate on different factors constituting Eqs. (5) which characterise behavior of neutrons in Oklo reactors.

2.1.1 Regeneration factor, η :

It is defined as number of neutrons produced per neutron absorbed in fuel. Definition of fuel is to some extent arbitrary. Though in Oklo uranium-235 was the only fissile isotope but we will define fuel as both uranium isotopes because both isotopes of uranium are chemically closely associated. Then regeneration factor is given by following equation.

$$\eta = \frac{\sigma_f^{25} \cdot \bar{\nu}_5}{(\sigma_a^{25} + \frac{N_{28}}{N_{25}} \sigma_a^{28})} \qquad (7)$$

where N_{25} is number density of uranium-235

N_{28} is number density of uranium-238

σ_f^{25} is fission cross-section of uranium-235

σ_a^{25} is absorption cross-section of uranium-235

σ_a^{28} is absorption cross-section of uranium-238

N_{28}/N_{25} varies with geological time as decay rates of two isotopes are different. Decay equations of two isotopes are

$$N_{28} = N_{28}^o \, \exp(0.153691 \, \tau)$$

$$N_{25} = N_{25}^o \, \exp(0.972156 \, \tau)$$

where N_{28}^o and N_{25}^o are number densities of two isotopes today and τ is geological time in units of billion years. It increases as we go in past. Relation abundance of two different isotopes is universally constant at any given time. Today $N_{28}^o/N_{25}^o = 137.85$. This implies

$$\frac{N_{28}}{N_{25}} = \frac{1}{137.85} \, \exp(-0.818465 \, \tau) \qquad (8)$$

From above equation we can see that uranium-235 abundance was more in past. Fig. (1) was plotted from above equation. Substituting above equation in (7) we get η as a function of geological time

2.1.2 Thermal utilisation factor, f :

Thermal utilisation factor is defined as the ratio of number of thermal neutrons absorbed per unit time in fuel to

the total number of thermal neutrons absorbed per unit time in reactor. For effectively homogenious reactor it is given by following equation.

$$f = (1 + \sum_{i=1}^{n} \frac{\sigma_a^i N_i}{\sigma_a^f N_f})^{-1} \tag{9}$$

where

σ_a^i is thermal neutron absorption cross-section of ith material in reactor zone.

N_i is number density of ith material in reactor zone.

σ_a^f is absorption cross-section of fuel $= \dfrac{N_{28}\sigma_a^{28} + N_{25}\sigma_a^{25}}{N_{25} + N_{28}}$

and $N_f = N_{28} + N_{25}$

From Eqs. (1), (2), (3) and (4) number densities N_i as a function of uranium content U%, geological time τ, neutron poison content x, and volume content of water in pores V%, can be evaluated. Substituting them in Eq.(9) we will get f(U%, τ, x, V%.

2.1.3 Resonance escape probability, p :

This factor takes care of the absorption of neutrons in epithermal energy range. Uranium is main absorber of epithermal neutrons and other materials are weak absorbers. Expression of resonance escape probability for every element is

$$p_i = \exp\left(- \frac{N_i}{\overline{\xi}\, \Sigma_{s/e}} \cdot I_{eff}\right)$$

$$I_{eff} = \int_{E_{th}}^{E_0} \frac{1}{1 + N_i \sigma_{a/e}^i / \Sigma_{s/e}} \; \sigma_{a/e}^i \; \frac{dE'}{E'} \qquad (10)$$

where

$\overline{\xi}$ is average logarithmic decrement

$$= \sum_{i=1}^{n} \sigma_{s/e}^i N_i \xi_i \Bigg/ \sum_{i=1}^{n} \sigma_{s/e}^i N_i$$

$\Sigma_{s/e}$ is total macroscopic epithermal scattering cross-section

$$= \sum_{i=1}^{n} N_i \, \sigma_{s/e}^i$$

$\sigma_{s/e}^i$ is microscopic epithermal scattering cross-section of ith element

$\sigma_{a/e}^i$ is microscopic epithermal absorption cross-section of ith element

I_{eff} is effective resonance absorption integral.

For weak absorbers in our reactor like uranium-235, Si, Al, Mg, Fe, Mn, K and O_2 above equation for each reduces to

$$p_i = \exp\left(- \frac{N_i}{\overline{\xi}\, \Sigma_s} \; I_{inf.dil.}\right) \qquad (11)$$

where $I_{inf.dil.}$ is infinite dilution resonance integral.
Epithermal absorption by hydrogen can be neglected.

For uranium-238 N_{28} $\sigma_{a/e}^{28}/\Sigma_{s/e}$ can not be neglected because of its large quantity in reactor zone. Experimentally determined values of integral in Eq. (10) are available for different values of $N_{28}/\Sigma_{s/e}$ [ ]. Following imperical relation can be used for integral.

$$I_{eff}(28) = \exp(0.46 \log(\Sigma_{s/e}/N_{28}) + 1.123)$$

$$p_{28} = \exp(-\frac{N_{28}}{\bar{\xi} \Sigma_{s/e}} I_{eff}(28)) \qquad (12)$$

Using equations (11) and (12) total resonance escape probability

$$p = p_{28} (\prod_{i=1}^{n} p_i) \qquad (13)$$

p is a function of U% , τ, x, V% .

2.1.4 Fast fission factor, ε :

It is a measure of neutrons born in U-238 fission by fast neutrons. In homogenious reactors it is quite close to one but in Oklo reactors fast effect is considerable because U-238/water ratio is quite high. Calculation for this factor is difficult without lot of assumptions. We assume that only elastic collision of fast neutrons with water brings it down below fast fission threshold of about 1 MeV and elastic collisions with other materials present in reactor do not alter fast fission probability by neutrons. Another assumption we make is that only inelastic collision with U-238 is possible and it brings down neutron below fission threshold. In another

words we have assumed for calculation of fast effect that Oklo
reactors only have homogenious mixture of U-238 and water. In
this case ε is given by following equation.

$$\varepsilon = (1 + 0.69\ N_{28}/N_w)/(1 + 0.563\ N_{28}/N_w) \qquad (14)$$

where N_w is number density of water.

This would mean ε is a function of τ, U% and V% .

2.1.5 *Diffusion area, L^2 :*

Diffusion area is related to mean square distance
travelled by neutron in thermal energy range. It is given by
following equation

$$L^2 = D_{th}/\Sigma_{a/th} \qquad (15)$$

where

$\Sigma_{a/th}$ is total thermal macroscopic absorption cross-
section $= \displaystyle\sum_{i=1}^{n} N_i\ \sigma_{a/th}^i$

and D_{th} is thermal diffusion coefficient

$$= 1/(3\ \Sigma_t(1-\bar{\mu}_o)(1- \frac{4}{5}\ \frac{\Sigma_{a/th}}{\Sigma_t} + \frac{\Sigma_{a/th}}{\Sigma_t}\ \frac{\bar{\mu}_o}{1-\bar{\mu}_o}))$$

Σ_t is thermal macroscopic total cross-section

$$= \sum_{i=1}^{n} N_i\ \sigma_t^i$$

$\bar{\mu}_o$ is average of cosine of scattering angle.

Equation (15) will give L^2 as a function of U%, τ, x and V%. In Oklo reactor medium scattering cross-section will have some variation in thermal range because of interaction between neutron and the molecular structure of medium or from interaction between neutron and the crystal structure of the medium. That is why calculated value of L^2 from Eq. (15) will be an underestimate.

2.1.6 *Neutron age, τ' :*

Age is related to mean square distance travelled by newly born fast neutron to thermal energy range. It is given by following expression:

$$\tau' = \frac{u-u_o}{\bar{\xi}} \left(3 \sum_{i=1}^{n} (N_i \sigma_{s/e}^i (1-\bar{\mu}_i)) \sum_{i=1}^{n} (N_i \sigma_{s/e}^i) \right)^{-1}$$

(16)

where

$\bar{\mu}_i$ is average cosine of scattering angle of ith element

$(u-u_o)$ is lethargy change during slowing down $= \ln \frac{E_o}{E}$. Value of E_o can be fixed at 2 MeV but value of E depends on temperature of the medium (E = kT) where k is Boltzmann constant.

From Eq. (16) we will get expression of τ' as a function of U% , τ, V% , thermal temperature of neutron.

2.1.7 *Modification of criticality equation :*

Major inadequacy in the four factor formula is omission of non-thermal fissions. In Oklo reactors moderation was not

very effective with the result that thermalisation of neutrons
was not complete. An important consequence of this is that
there was significant contribution to the multiplication from
fission in U-235 caused by absorption of epithermal neutrons.
Oklo reactors being a slightly enriched system (enrichment
near 3%) this becomes all the more important. Here we will
develop dependence of neutron multiplication factor on epithermal
fission of U-235.

In order to establish a physical picture of the factors
included, neutron cycle given in Fig. (12) is developed. Let
at point A one thermal neutron is absorbed. As a result ηf
fission neutrons will be produced as shown by line AB. To these
fast neutrons x fission neutrons will be added by the epithermal
fission of U-235, as indicated by line DB, so that the total
number of fast neutrons at B is $(\eta f + x)$. As a consequence of
fast fission effect in U-238 total number of fission neutrons
available for slowing down is $(\eta f + x)\varepsilon$. Process of slowing
down is arbitrarily divided into two parts, first from C to D,
taken to be 0.625 eV, where radiation capture in U-235 is
significant, and then from D to E, where the energy is thermal.
The number of neutrons reaching D is equal to $(\eta f + x)\varepsilon P_1 p'_{28}$,
where P_i is the non-leakage probability of neutrons with ener-
gies above D and p'_{28} is the neutron resonance escape probability
for uranium-238 in the same energy range. In slowing down from
D to E, the following additional factors are introduced: P_2,
the non-leakage probability for neutrons in the energy range D
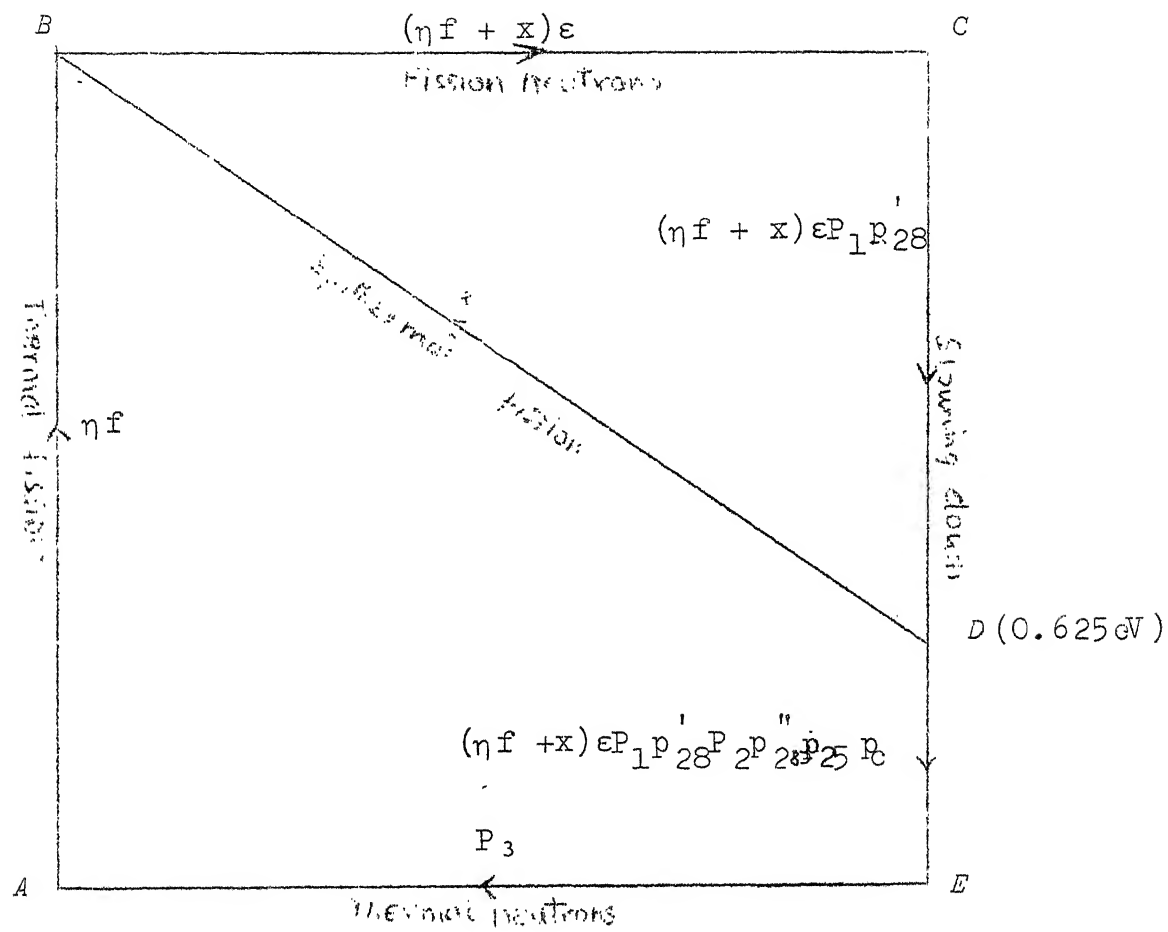
Fig. 12: Modified neutron cycle.

D to E; p_{25}, capture escape probability for U-235; and p_c, the probability that neutron while slowing down will escape capture in various materials other than fuel. Finally, introduction of P_3, the non-leakage probability of thermal neutrons completes the cycle. The effective multiplication factor is given by

$$K_{eff} = P_1 P_2 P_3 (\eta f + x) \varepsilon \, p'_{28} \, p''_{28} \, p_{25} \, p_c \quad (17)$$

Since p_{25} is the capture escape probability for U-235, the fraction of neutrons reaching the point D which undergo epithermal absorption in U-235 is $(1 - p_{25})$. Consequently, the number x of fission neutrons produced by this process is

$$x = \eta_{25}(1 - p_{25}) \left[(\eta f + x) \varepsilon \, P_1 \, p'_{28} \right]$$

where η_{25} is the number of neutrons produced per epithermal neutron absorbed in U-235. From above relation it follows that

$$(\eta f + x) = \eta f [1 - \varepsilon \, P_1 \, p'_{28} \, (1 - p_{25})]^{-1}$$

Substituting it into Eq. (17) yields

$$K_{eff} = P_1 P_2 P_3 \eta \, f \, \varepsilon \, p'_{28} \, p_c \, \beta \, p''_{28} \quad (18)$$

where

$$\beta \equiv p_{25} \left[1 - \varepsilon \, P_1 \, p'_{28} \, \eta_{25} \, (1 - p_{25}) \right]^{-1} \quad (19)$$

With $P_1 P_2 = \exp(-\tau' B^2)$ and $p'_{28} p''_{28} = p_{28}$ Eq.(18) reduces to

$$K_{eff} = \frac{\eta\, f\, \varepsilon\, p_{28}\, p_c\, \beta}{1 + L^2 B^2} \ \exp(-\tau' B^2) \qquad (20)$$

Equation (20) is quite similar to original criticality equation (5) except for factor β, which takes care of epithermal fission and capture by U-235. For evaluation of β, P_1 and p'_{28} have to be evaluated. η_{25} is constant. p_{25} and ε have been formulated in Section 2.1.4 and 2.1.5. P_1 is non-leakage probability for energy range 2 MeV to 0.625 eV and is given by

$$P_1 = \exp[-\tau'(2\,MeV - 0.625\,eV)\, B^2]$$

where expression for $\tau'(E_0 - E)$ is given in Section 2.1.7.

Evaluation of p'_{28}, neutron resonance escape probability in energy range B to D for U-238 is difficult to calculate. It is possible to write $p'_{28} = p^n_{28}$, where n is imperically derived constant. It is found that best fit to the data is obtained by taking n = 0.5 [37]. This would imply,

$$p'_{28} = p^{0.5}_{28} \qquad (21)$$

where evaluation of p_{28} is elaborated in Section 2.1.4.

Now we are in a position to express K_{eff} as a function of uranium constant U% , geological time τ, poison constant x, volume content of water in rock pores V% , content of water structurally bound with rock, geometric buckling B^2, and

temperature of thermal neutron, T_n. Temperature dependence of K_{eff} is more complicated and is dealt in next section.

2.2 Temperature dependence of K_{eff} :

Major dependence of K_{eff} on temperature comes from dependence of microscopic absorption and fission cross-sections of elements in reaction zone on temperature as well as temperature dependence of resonance integrals. For Oklo, expansion of unbound water in pores, m in equation (4), also is significant. Kind of formalism we have adopted in Section 1.6 does enable us to take into account expansion of water in pores, which would be in supercritical state. To take this into consideration would require knowledge of volume of pores in Oklo rocks, V%, which is not available. As we are keeping V% as an open variable this drawback does not inhibit us from studying criticality condition accurately. Study of reactor behavior after reaction has began will not be complete unless expansion of water is taken into account with rise of temperature as fission proceeds.

2.2.1 Cross-section dependancie on temperature :

Absorption cross-section of all the elements present in reactor zone except U-235 have been assumed to behave according to 1/v law in the thermal region. Averaged thermal cross-section for these elements is given by

$$\sigma(th) = \frac{\sqrt{\pi}}{2} \left(293.6/T_n \right)^{1/2} \sigma(2,200)$$

$\sigma(2,200)$ is cross-section for neutron with speed 2200 m/sec and is given in appendix (I) for all elements of concern. T_n is neutron temperature on the Kelvin scale. $\sqrt{\pi}/2$ is a correction for Maxwell distribution of thermal neutrons.

For U-235 Westcott correction have been used to evaluate effective absorption and fission cross-sections. Effective cross-section according to Westcott's convention is given by

$$\sigma_{eff} = [\, g(T_n) + r\, S(T_n)]\, \sigma\,(2,200)$$

where $g(T_n)$ is a measure of the departure of cross-section from $1/v$ dependence average over Maxwellian component and can be approximated [38] as

$$g_a^{25} = -\,0.7245 \times 10^{-4}\,(T_n - 100) + 0.952$$

$$g_f^{25} = -\,0.10333 \times 10^{-3}\,(T_n - 100) + 0.9495$$

for absorption and fission, respectively. $S(T_n)$ is the contribution to the total reaction rate from the epithermal part of the spectrum minus Maxwellian distribution and can be approximated as

$$S_a^{25} = 0.105 \times 10^{-3}\,(T_n - 100) + 0.1185$$

$$S_f^{25} = 0.32 \times 10^{-4}\,(T_n - 100) + 0.014$$

for absorption and fission, respectively.

Parameter r is used to normalise the relative contribution of the epithermal and Maxwellian component of neutron spectrum. Value of r was estimated by irradiating different samples of Oklo rock in Triton reactor by French study group[39] to be between 0.10 to 0.25.

T_n is neutron temperature. In Oklo neutron temperature is more than $150^{\circ}C$ in excess of thermal temperature of melieu. This is because of high thermal absorption and incomplete moderation. Relation between temperature of melieu and neutron temperature is

$$T_n = T_m (1 + 0.46 \quad) \qquad 1 < \text{mass number} < 25$$

$$T_n = T_m (1 + 0.30 \quad) \qquad 25 < \text{mass number}$$

where

$$= \frac{4 \; \Sigma_a \; (K \; T_m)}{\bar{\xi} \; \Sigma_s}$$

$\Sigma_a (K \; T_m)$ is total macroscopic absorption cross-section at melieu temperature $= \sum_{i=1}^{n} \sigma_a^i (K \; T_m) \; N_i$

Σ_s is total epithermal scattering cross-section

$$= \sum_{i=1}^{n} N_i \; \sigma_{s/e}^i \cdot$$

and $\bar{\xi}$ is average logarithmic decriment define in Section 2.1.4.

2.2.2 Resonance integral temperature dependence :

In the formulation developed above neutron interaction process has been idealised as neutron impinging upon a scattering or absorbing atom which is at rest. Target atom in actuality has a thermal motion. With respect to the average position of the target atom neutron will seem to have spread out energy distribution. It can be shown for a Maxwellian distribution of target nucleus velocities and $1/v$ cross-section, that Doppler effect makes no difference in the effective cross-section of the interaction. Only for resonances this effect becomes important. It lowers Breit-Wigner resonance peak height and spreads it but area under peak is not changed, so that for dilute elements or elements having small RI there is negligible change in resonance integral with temperature. However for U-238, widening of the region of strong capture would appreciably increase absorption in skirts of resonances. From experimentally arrived values of resonance integral for U-238 with temperature, given in ANL 5800 [38] following analytic form is constructed

$$I_{eff}(T) = 7.46166 \times 10^{-3} (T_m - 27) + \exp[0.46 \log(\frac{\Sigma s/e}{N_{28}}) + 1.123]$$

where T_m is in $^{\circ}C$ and is temperature of milieu.

Now formulation of K_{eff}, for study of natural reactors, as function of uranium content U% , loose water content V% , poison content x, geological time τ, geometric buckling B^2, and

temperature of milieu is complete. In Appendix (II) is given
constructed computer programme for study of criticality
conditions.

Variation of above given six parameters, within cons-
traints set in chapter one, can be investigated to find out
situation when K_{eff} is one or more. Effect of temperature on
four factors and K_{eff} can be studied. Also other features of
natural reactors for comparative study of reactor systems
can be evaluated from constructed model.

2.3   Results :

2.3.1   Parametric dependence of Neutron temperature :

In Fig. (16) is plotted variation of neutron temperature
with temperature of milieu for different uranium content. For
milieu temperature of $200^{o}C$ neutron temperature is $270^{o}C$ for
$U\% = 24\%$. The difference is large enough indicating that
distinction between milieu temperature and neutron temperature
is crusial for modelling at Oklo reactors. This difference is
high because of large thermal absorption cross-section and less
slowing down power.

With increase in uranium content neutron temperature
rises because milieu content decreases as is expected from
Eq. (2). This leads to less water content as is evident from
Eq. (4). Net effect is worsening of slowing down power of
materials in Oklo rock leading to high degree of non-moderation.
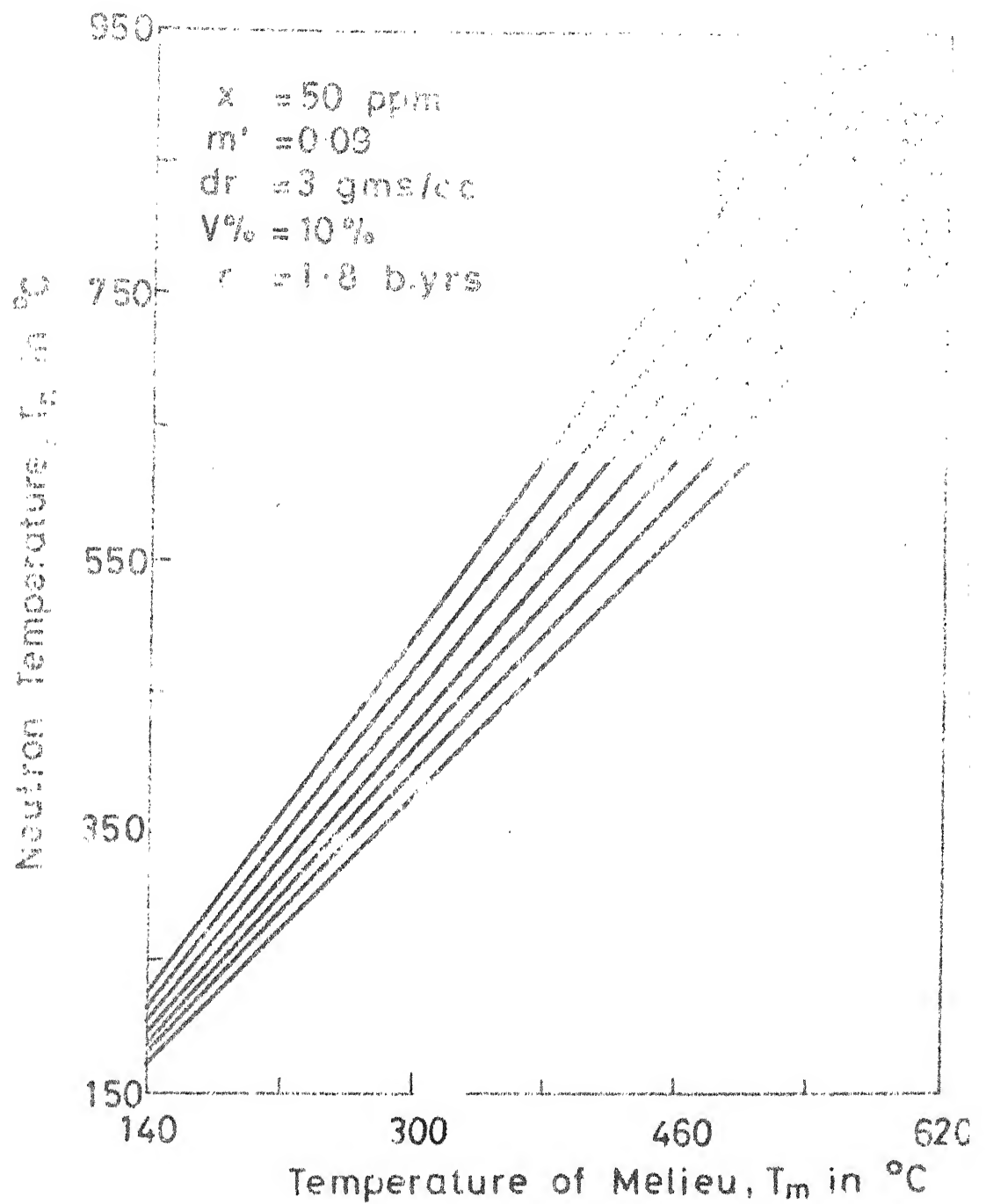
Fig.16 Variation of Neutron temperature with Melieu temperature

Another significant parameter which dictates neutron temperature
is water content itself. For lower milieu temperature the
effect of change in V% is not much but for higher temperatures
neutron temperature becomes very sensitively dependent on V%.
It also has a dependence on B^{10} equivalent poison content, x
as strong thermal capture implies large gap between milieu
temperature and neutron temperature because of quicker disappea-
rance of thermalised neutrons. Understanding of neutron tempera-
ture behavior is important for understanding parametric dependence
of factors which characterise behavior of neutrons in reactor.

2.4.2 *Parametric dependence of Neutronic factors :*

Table (3) gives variation of different factors which
characterise behavior of neutrons with different parameters.
With rise in water content, y, neutrons are slowed down more
efficiently. Age, τ', and contribution of epithermal fission
of U-235, β, decreases but resonance escape probability increases
Also neutrons are brought down below fast fission threshold
quickly, decreasing fast fission factor, ε. In thermal region
parasitic capture increases owing to which thermal utilisation,
f, and diffusion area, L^2, decreases. For more content of water
neutron temperature is less and temperature dependence of fission
and absorption cross-sections of two isotopes of uranium is such
that η decreases slightly. For low temperature fission cross-
section of U-235 decreases faster with temperature than aggregate
of macroscopic absorption cross-section of fuel. Decrease in
τ' and increase in p and η increases K_{eff} but decrease in f, L^2

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TABLE - 3

V%	U%	X	Y	ε	η	p	f	β	k_inf	L²	τ'	k_eff
8.0	16.0	2.7643	.3626	1.0206	1.8499	.6737	.7372	1.0318	.9376	.6661	5.851	.9612
10.0	16.0	2.7643	.3910	1.0192	1.8497	.6681	.7320	1.0301	.9495	.6261	5.281	.9723
12.0	16.0	2.7643	.4135	1.0180	1.8495	.7010	.7268	1.0285	.9592	.5903	4.793	.9814
8.0	20.0	2.0753	.2778	1.0261	1.8504	.6437	.7854	1.0397	.9600	.8388	9.118	.9882
10.0	20.0	2.0753	.3006	1.0244	1.8502	.6597	.7806	1.0376	.9759	.7878	8.193	1.0036
12.0	20.0	2.0753	.3235	1.0228	1.8500	.6739	.7758	1.0358	.9894	.7422	7.406	1.0164
8.0	24.0	1.6159	.2213	1.8510	1.0318	.6155	.8216	1.0475	.9658	.9927	13.090	.9975
10.0	24.0	1.6159	.2403	1.0297	1.8508	.6330	.8171	1.0452	.9856	.9312	11.706	1.0172
12.0	24.0	1.6159	.2592	1.0278	1.8506	.6486	.8127	1.0430	1.0025	.8764	10.537	1.0338
8.0	28.0	1.2878	.1809	1.0378	1.8516	.5884	.8498	1.0552	.9607	1.1258	17.758	.9947
10.0	28.0	1.2878	.1972	1.0351	1.8514	.6075	.8456	1.0526	.9844	1.0545	15.797	1.0189
12.0	28.0	1.2878	.2134	1.0328	1.8511	.6245	.8415	1.0501	1.0048	.9912	14.154	1.0393
8.0	32.0	1.0418	.1507	1.0439	1.8522	.5619	.8725	1.0625	.9479	1.2371	23.109	.9829
10.0	32.0	1.0418	.1649	1.0408	1.8519	.5828	.8686	1.0598	.9756	1.1566	20.439	1.0119
12.0	32.0	1.0418	.1791	1.0381	1.8516	.6013	.8648	1.0573	.9994	1.0855	18.221	1.0364

$d_r = 3$ gm/cc, $m' = 0.09$, Tt = 200°C, x = 50 ppm, $B^2 = 10$ m^{-2}, T = 1.8 billion years.

and ε decrease it. Owing to these push and pull it is maximum at some optimal point and decrease on both sides. Content of water, y, is so less that this maxima is not reached in region of interest hence K_{eff} shows an monotonic increase with water content rise.

With increase in uranium content, U%, content of milieu decreases implying less parasitic capture in thermal region increasing f and L^2. In epithermal region less milieu content make slowing down inefficient thus increasing τ'. Rise in U-238 content decreases p but increases ε slightly. More U-235 content increases β. η increases with increase in uranium content because of rise of neutron temperature. K_{eff} increases with rise in U% but for considerably high U% it decreases owing to less slowing down because of decrease in milieu and water content which eventually makes decrease in p significant. Effect of change in geological time is not shown in Table (3).

With increase in geological time enrichment is more increasing η. Rise in U-235 content would lead to more thermal neutron absorption decreasing L^2 but f would increase as relative increase in fuel absorption would more than increase in total absorption in thermal region. Increase in geological time decrease milieu content decreasing water content thus lowering resonance escape probability as neutron flux in epithermal region will increase. Because of non-moderation ε also increases as pulling down of neutrons below fast fission threshold becomes inefficient. Non-moderation would increase τ' as well. Net effect of K_{eff} is because of increase in fuel enrichment which increases it.

Effect of change in poison content is also not shown in Table (3). It will only effect thermal factors f and L^2. Lowering them both with increase in poison content.

B^2 is a geometric property and not intrinsic nuclear property. It has no effect on different factors. Increase in B^2 would increase leakage of neutrons out of reaction site. Thus will lower K_{eff}.

2.3.3 Dependence of neutronic factors on temperature of milieu :

Table (4) illustrates temperature dependence of different factors of interest for U% = 24% , V% = 10% , m' = 0.09, x = 50 x 10^{-6}, T = 1.8 b_y and d_r = 3. Relative content of water, y, decreases because of expansion of water in pores. Increase in temperature decreases absorption cross-section of materials in reaction site. This leads to decrease in parasitic capture in thermal range increasing f and L^2. Decrease in slowing down power because of decrease in y explains increase in ε, τ and β and decrease in p. Behavior of η is puzzling, it increases reaching maximum around 340°C and then decreases. This is because of non 1/v temperature dependence of fission and absorption cross-sections of both isotopes of uranium. But for our range of interest from 150°C to 300°C it increases. K_{eff} also increases till 240°C and then decreases. This is a significant result contrary to most of thermal man-made reactors where temperature coefficient of reactivity is invariably kept negative. Distinct feature of Oklo reactors is that there is lot of parasitic

TABLE 4

T_m (°C)	Y	f	η	ε	p	K_{inf}	L^2	τ'	β	K_{eff}
140	0.246	0.8052	1.8455	1.02905	0.6409	0.98003	0.8906	11.43	1.0448	1.01140
160	0.244	0.8064	1.8476	1.02925	0.6382	0.98247	0.9044	11.52	1.0449	1.01393
180	0.242	0.8134	1.8493	1.02946	0.6357	0.98431	0.9179	11.61	1.0450	1.01584
200	0.240	0.8171	1.8508	1.02966	0.6330	0.98562	0.9312	11.71	1.0452	1.01719
220	0.238	0.8205	1.8510	1.02987	0.6303	0.98645	0.9443	11.81	1.0453	1.01805
240	0.235	0.8238	1.8520	1.03009	0.6276	0.98685	0.9573	11.91	1.0454	1.01845
260	0.234	0.8269	1.8537	1.03030	0.6249	0.98684	0.9701	12.01	1.0455	1.01844
280	0.232	0.8299	1.8541	1.03052	0.6221	0.98647	0.9828	12.12	1.0456	1.01804
300	0.231	0.8327	1.8545	1.03074	0.6193	0.98576	0.9954	12.23	1.0457	1.01728
320	0.229	0.8353	1.8547	1.03097	0.6165	0.98472	1.0080	12.34	1.0458	1.01619

$U\% = 24\%$, $V\% = 10\%$, $m' = 0.09$, $x = 50$ ppm, $\tau = 1.8$ b.yrs. $d_r = 3$ gm/cm³

$B^2 = 10$ m^{-2}.

capture in thermal range which has $1/\sqrt{T}$ dependence over
temperature. So at lower temperatures f quickly increases
and corresponding increase in L^2, which negatively contributes
to K_{eff}, is ineffective because of large geometric buckling
and small absolute value of L^2. Because of small change in
quantity of water, y, at lower temperature decrease in resonance
escape probability is not much to counter increase in f. As
the temperature is increased decrease in p eventually matches
increase in f thus bringing down K_{eff} eventually. Another
points to note at this stage is that temperature dependence of
resonance integral of materials other than uranium was not
consider owing to small values of there resonance integrals.
In actuality there resonance integral will increase with
temperature, increasing rate of decrease of p but the relative
significance of contribution of rest of materials to p is much
less than that of U-238 whose temperature dependence was
taken into account.

2.4.4 Neutron balance table :

To aid establish physical picture of chain reaction for
natural reactors of Oklo, neutron balance sheet is made and is
given in Table (5). Parameters were chosen to give $K_{eff} = 1$.
B^2 was taken 10 m^{-2} for reactors of size of reactor zone 1 and 2.
U% was taken 24% which is about average content of reactor
zone 1. Content of chemically bound water was taken to be 9%,
content of loose water 10% , poison content 50 ppm and tempera-
ture when reaction began to be 200°C. Time when K_{eff} is one was

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TABLE 5

NEUTRON BALANCE SHEET

| 10000 THERMAL NEUTRONS ABSORBED IN REACTOR |

| 9 THERMAL NEUTRONS LEAK OUT OF REACTOR |

| 1917 PARASITIC CAPTURE OF THERMAL NEUTRONS BY MELIEU |

| 10009 NEUTRONS THERMALISE IN REACTORS |

| 8083 THERMAL NEUTRONS ABSORBED IN FUEL |

| 96 NEUTRONS LEAK OUT BETWEEN RESONANCE AND THERMAL ENERGIES |

| 419 NEUTRONS ADDED BY EPITHERMAL FISSION OF U-235 |

| 2053 NON-PRODUCTIVE CAPTURE OF THERMAL NEUTRONS BY FUEL |

| 9696 NEUTRON ESCAPE RESONANCE CAPTURE |

| 6030 THERMAL NEUTRONS LEADING TO FISSION |

| 5614 NEUTRONS CAPTURED IN RESONANCE REGION |

| 15310 NEUTRONS ENTER RESONANCE REGION |

| 14894 FAST NEUTRONS PRODUCED BY FISSION |

| 24 NEUTRONS LEAK OUT BEFORE ENTERING RESONANCE REGION |

| 15334 FAST NEUTRONS BY THERMAL AND FAST FISSION |

| 440 ADDED BY FAST-FISSION OF U-238 |

found out to be 1741.22 million years and path of neutron cycle was followed to give this balance table. With the help of this table one can comparitively study position of natural reactors with respect to man-made reactors.

Neutron produced by fast fission of U-238 is 5% of neutron produced by thermal fission of U-235. 4.73% of fast neutrons are produced by U-235 epithermal fission. 19% of thermal neutrons are captured by milieu and 20.5% are non-productively captured by fuel to produce U-236 and Pu-239. Only 0.8% neutrons leak out of reactor from the time fast neutron is born to the time it is absorbed in thermal region after slowing down. Parasitic capture of thermal as well as epithermal neutrons is significantly high in Oklo reactors as compared to reactors made under intended care of man. Leakage of neutrons is extremely small compared to modern slightly enriched reactors where 5 to 15% of neutrons leak out.

Table (6) lists some reactor physics features of Oklo reactors for same combination of parameters as in neutron balance sheet. Average logarithmic energy decrement is quite near that of D_2O which is 0.57. Also number of collisions required to slow down neutrons is close to D_2O which is 35. Slowing down time for D_2O moderater is 0.5×10^{-4} sec for Oklo it is 0.476×10^{-4} sec. But moderating power is much higher than that of pure D_2O which is 0.18. This is because total number density of elements in Oklo reactors is 0.2×10^{24} where as for D_2O of density 1.1 gm/cm³

TABLE 6

SOME REACTOR PHYSICS FEATURES OF OKLO REACTORS

Average logarithmic energy decrement, $\bar{\xi}$	=	0.5643
Number of collisions before neutron slows down	=	32
Moderating Power, $\overline{\xi \Sigma_s^{ep}}$	=	0.59
Moderating ratio, $\overline{\xi \Sigma_s^{ep}} / \Sigma_a^{th}$	=	3.79
Enrichment of fuel, e	=	2.9283%
Total number density of uranium, Nu	=	0.625×10^{22}
Total number density of all materials	=	0.2×10^{24}
Macroscopic thermal absorption cross-section, Σ_a^{th}	=	0.15567 cm^{-1}
Macroscopic epithermal scattering cross-section, Σ_s^{ep}	=	1.04562 cm^{-1}
Thermal Macroscopic transport cross-section	=	70.993 cm^{-1}
Thermal diffusion length, $\sqrt{L^2}$	=	0.9781 cm
Slowing down length, $\sqrt{\tau'}$	=	3.4 cm
Migration length, $\sqrt{(L^2+\tau')}$	=	3.535 cm
Thermal prompt neutron life time	=	0.6814×10^{-2} sec.
Slowing down time	=	0.47565×10^{-4} Sec.

$m' = 0.09$, $d_r = 3$ gm/cc, $V\% = 10\%$, $U\% = 24\%$, $T_m = 200^{\circ}C$,

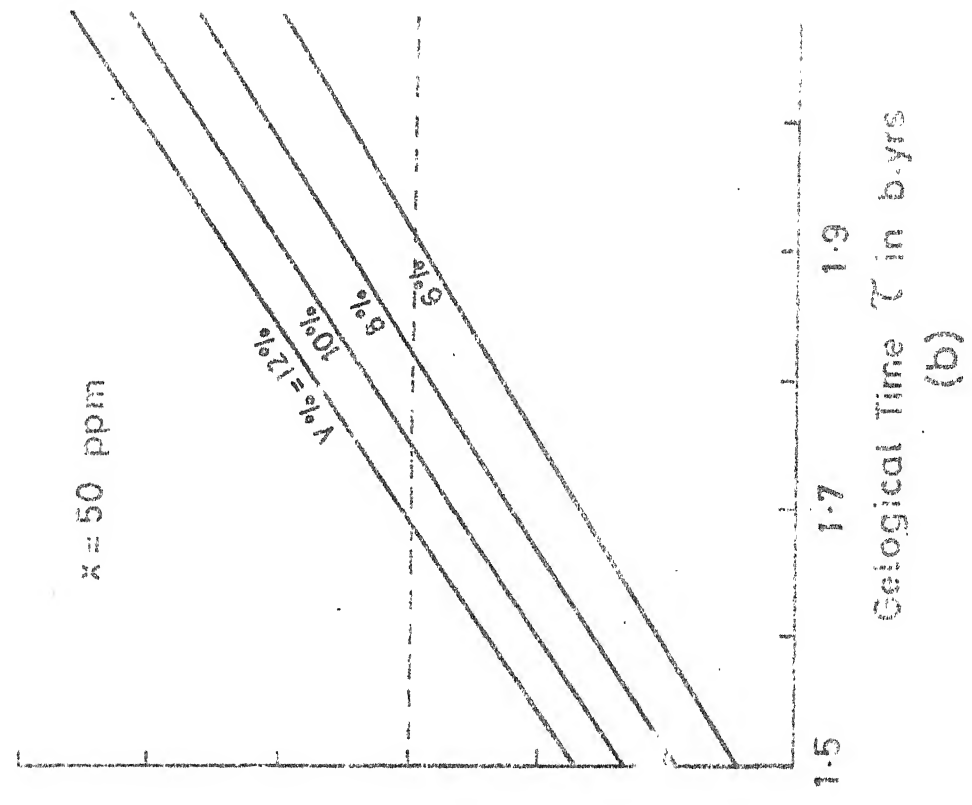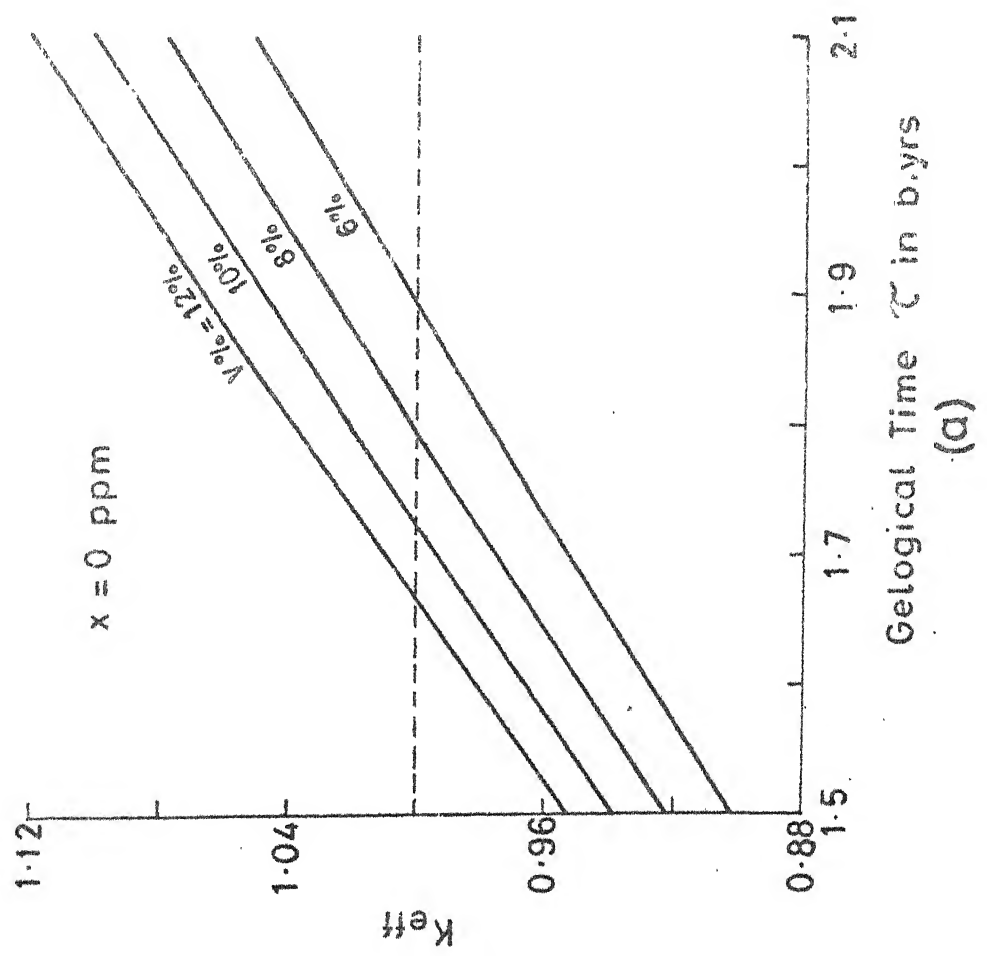$B^2 = 10$ m^{-2}, $x = 50$ ppm, $\tau = 1.74122$ billion years.

it is 0.0334 x 10^{24}. Because of high number density in Oklo rock slowing down length is only 3.535 cm where as even for pure water moderator it is 5.568 cm. Thermal diffusion length is considerably small because of high thermal macroscopic transport cross-section. Thermal macroscopic absorption cross-section for Oklo is very close to that of Iodine of density 4.93 gm/cm^3 which has macroscopic absorption cross-section 0.164 cm^{-1}. Thermal prompt neutron life time is less than that of heavy water but more than that of Beryllium.

2.3.5 *Study of criticality conditions :*

Now we go over to reflect on parametric conditions under which Oklo reactors could have become critical.

In Fig. (17a, b, c,,d) is plotted K_{eff} as a function of geological time, τ, for different volume fraction of loose water in rock pores. a, b, c, d graphs are for poison content of 0 ppm, 50 ppm, 100 ppm and 150 ppm, respectively. Sadimentary assemblage of 24% uranium by weight of the size of reaction zone 1 and 2 could have become critical as late as 1.67 byears ago there was no poison and loose water occupied 12% volume of rock. If poison content was equivalent to 150 ppm of B^{10} and V % = 6% then reactors could'nt have worked after 1.94 billion years back. At $200^{\circ}C$ of milieu temperature and m' = 0.09 in rock of density 3 gm/cm^3 possible range of time in which Oklo reactors could have worked by latest is 1.67 billion years. At any situation in this range assemblage could have become critical

(a)

$x = 0$ ppm

Vol = 12%
10%
8%
6%

Gelogical Time τ in b.yrs

(b)

$x = 50$ ppm

Vol = 12%
10%
8%
6%

Gelogical Time τ in b.yrs

$U\% = 24\%$
$m' = 0.09$
$B^2 = 10 \ m^{-2}$
$T_m = 200°C$
$dr = 3 \ gm/cc$
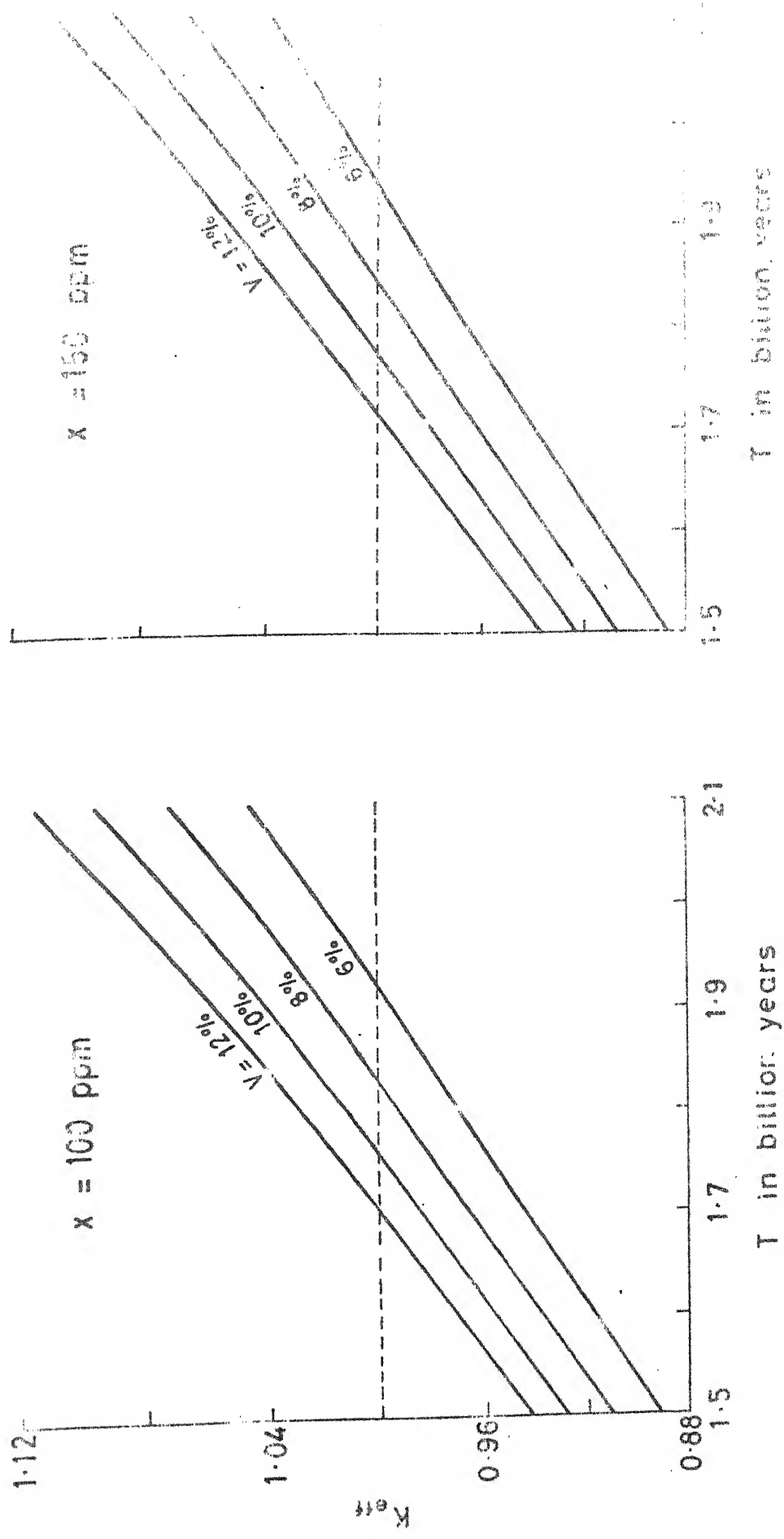
Fig.17(c)(d) Variation of effective Neutron multiplication factor with

in below given period

for no   poison   τ :   1.67 to 1.795 billion years
for 50   ppmℭ   τ :   1.685 to 1.81 billion years
for 100 ppm   τ :   1.7 to 1.83 billion years
for 150 ppm   τ :   1.716 to 1.845 billion years

Increase in milieu temperature would shift these curves slightly to the left side of graph till about 240°C and then again shift them to right at slightly lower rate if temperature was still higher. Change in limits of periods would for lowest temperature 160°C shift about 0.015 billion years and for optimal temperature of about 240°C it will shift left by about -0.012 billion years and then again boundaries would shift towards right aquiring almost above plotted position if temperature of milieu is around 300°C. τ is not very sensitive to temperature.

Notisable feature of Fig. (17) is sensitivity of geological time to water content. In Fig. (18) is plotted K_{eff} with net water content per unit UO_2, Y, for different uranium content U% , at 1.8 billion years ago. In making this plot Y was taken as an open variable disregarding formalism for it developed in Section 1.6.

For y = 0.5 assemblage could have become critical for even 16% uranium concentration by weight at time 1.8 billion years ago. It would have been critical with lesser uranium concentration before 1.8 billion years ago. All the six fossiles
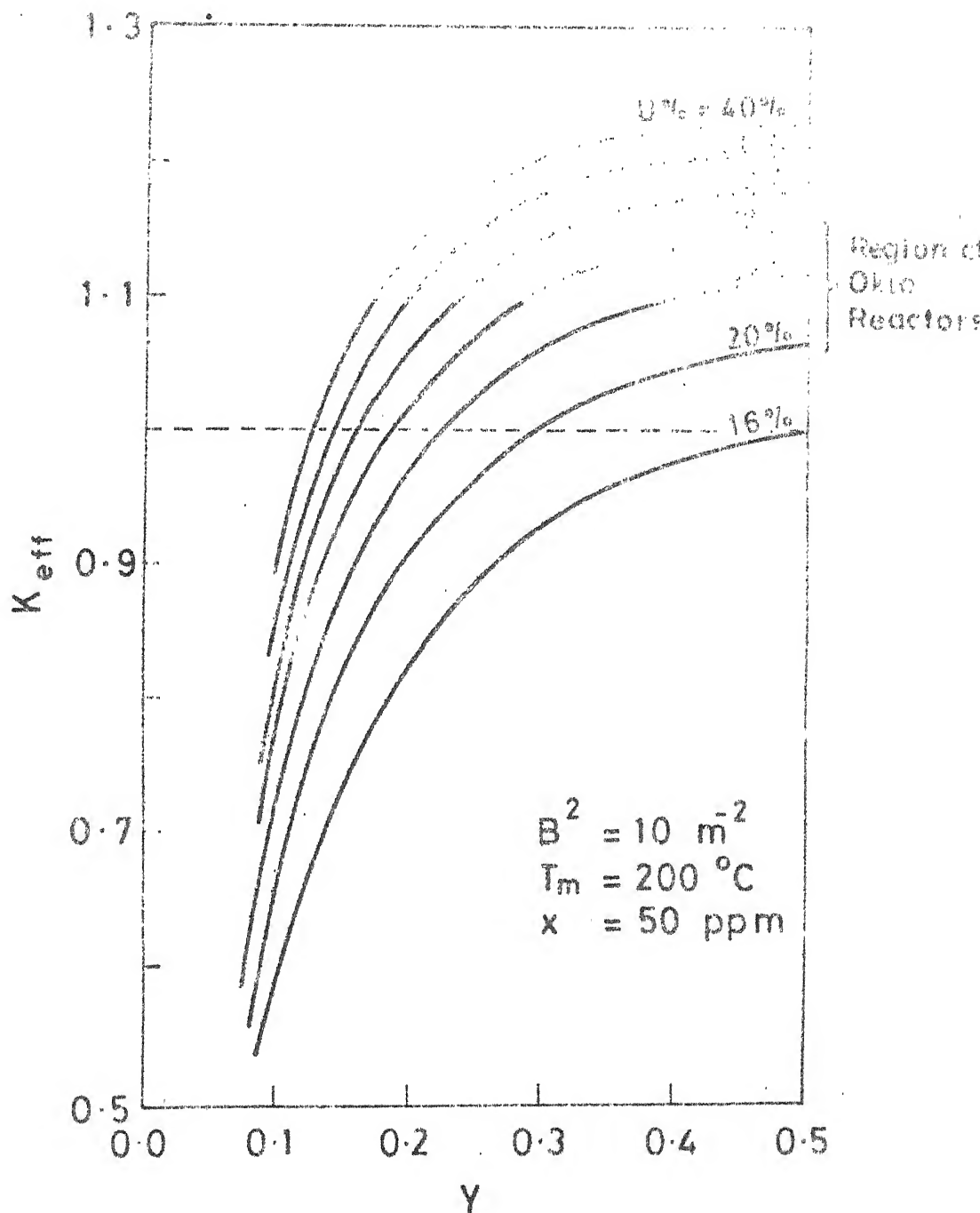
Fig. 18 Variation of effective Neutron
multiplication factor with net water
content

enearthed at Oklo have average relative weight content of uranium more than 20% . If reaction began 1.8 billion years ago with U% = 16% and y = 0.5 then how they have such high content can only be explained by assuming 4% to 12% more deposition took place after reaction began so that at the closing of chain reaction reactors had 20% to 28% uranium. If no significant deposition of uranium occured then for τ = 1.8 billion years, y should be less than 0.3. For τ greater than 1.8 billion years y would be even lesser. Hence to satisfy criticality requirement of Oklo V% need not be greater than 12% for d_r = 3 (from Fig. (15c and 15d).). So there was no shortage of water at Oklo when the reaction had to began only if our assumption that volume content of pores was between 8% to 12% is right. It is quite likely that Oklo rock lying at the base of main slippage dome in precambrian B orogeny had developed many microfractures to hold 8% to 12% water.

For smaller values of y reactor could have began earlier than 1.8 billion years. As Fig. (17d) indicates it would have at the earliest began by 1.85 billion years even if poison content was very high 150 ppm. If parameter temperature is increased it would pull up curves of Fig. (18) in the beginning but as temperature is increased more these curves will be pushed down. As K_{eff} does not have monolithic dependence on temperature the effect of temperature would not be very pronounced.

Fig. 19 Variation of Geomatric buckling with Uranium content
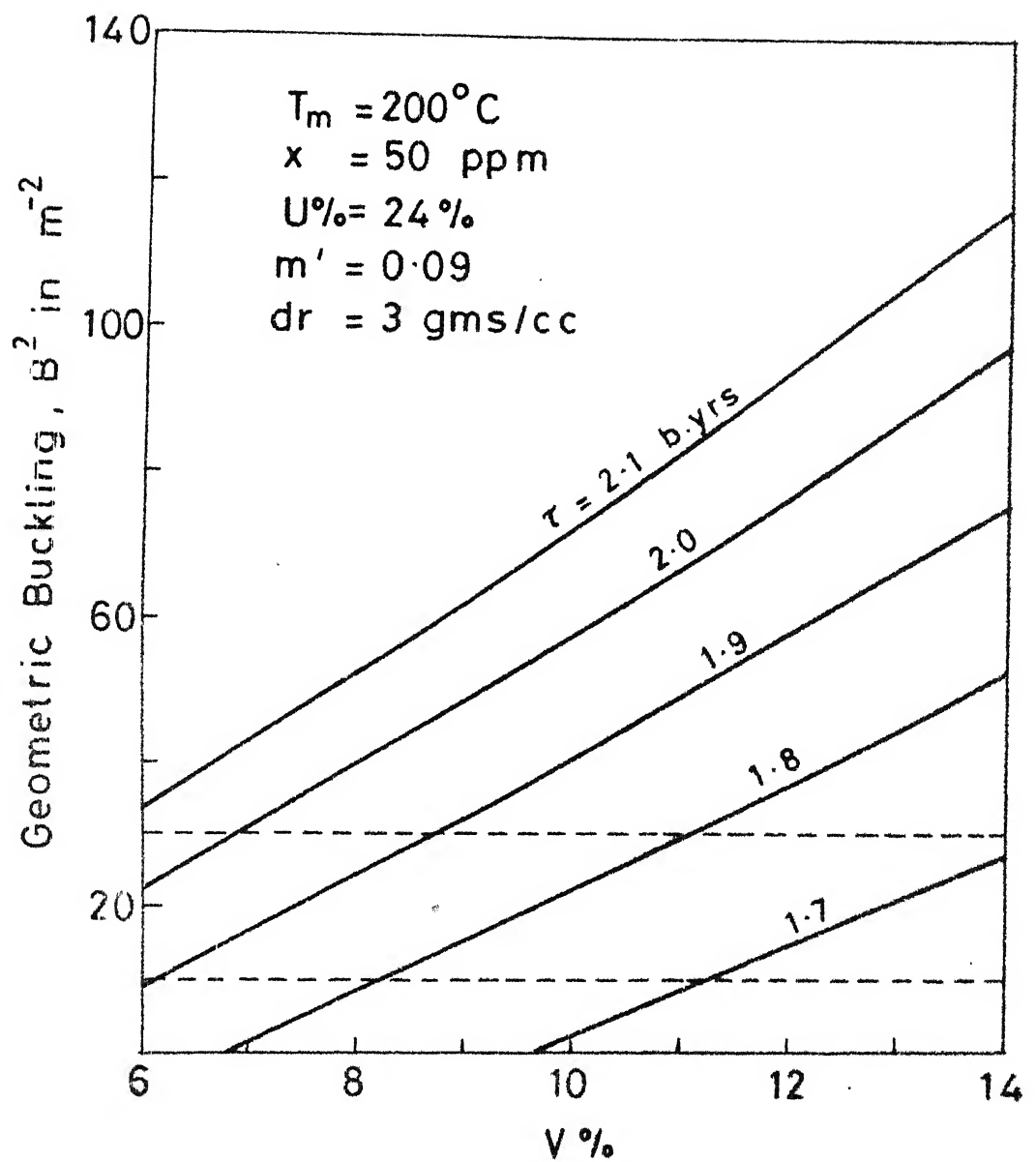
Fig. 20 Variation of Geomatric Buckling with loose water content

average of age of primary mineralisation was suggested to be near 1.9 billion years. Then it must have taken couple of tens of million years for sediments to reach such depth that desilicanisation was possible which was essential for reconcentration of uranium to take certain privilaged zone to criticality. Also conclusion from Fig. (17a,b,c,d) suggest age of start of reaction could not have been before 1.84 billion years. In this light it looks likely from Fig.(20) that porosity fraction of rock must be 8% or more as poison content could'nt have been very much less than 50 ppm. This sets lower limit to porosity. Upper limit to loose water content was infered from Fig. (15c and d) and Fig. (18) to be 12%. So our assumption that loose water content was between 8% to 12% is substantiated as necessary condition for possibility of reaction. Volume fraction lower than 8% is not compatible for establishment of criticality and greater than 12% is unnecessary.

Figure (19) gives plot of B^2 with respect to U% for different geological time for V = 10% and poison 50 ppm. This graph can be seen in the light of desilicanisation process in which uranium began concentrating. Desilicanisation process can be seen as motion along x-axis. From present uranium content of reactor fossiles one can infer than non of reactor began operating before reaching uranium concentration of about 20% by weight during desilicanisation. Limit 20% is set by accepting that upto 4% uranium could have got deposited after

reaction began. This would imply from Fig. (19) that biggest reaction could'nt have began some 1.79 billion years ago for $V = 10\%$. For higher V this upper limit shifts down and for lower V this limit shifts up. For reaction zone 3, 4, 5 and 6 B^2 was considerably higher upto 20 m^{-2} and these reactors could have worked around 1.79 billion years. Only when U% reached around 24%. For a situation when all the six reactors worked simultaneously we can conclude that either U% in smaller reaction zones should have been more than that of largest by 4% or that loose water content was more in smaller zones than in bigger.

For $V = 10\%$ smaller reactor zones with $U = 24\%$ could have only become critical before 1.78 billion years ago. This sets lower limit to time of their working. But it V%= 12% (from Fig.(20)) this lower limit shifts to about 1.76 billion years. If reactors were working simultaneously than the period in which they worked is between 1.81 to 1.74 billion years with lesser water content in larger zones and more water content in smaller zone.

If process of desilicanisation occured in very short time span compared to geological time considered than bigger reaction zones could be critical as late as 1.74 billion years for U% = 23.6% (from Fig.(19)). And for $V = 12\%$, we can say from Fig.(20) that bigger zones could have become critical as late as 1.68 billion years. This gives another picture of smaller zones becoming critical first around 1.81 billion years

to 1.76 billion years and later larger zones becoming critical upto as late as 1.68 billion years with $V = 12\%$ and poison 50 ppm. If we take B^2 for intermediate zones to be 15 m^{-2} then range for such reactors become 1.81 billion years to 1.73 billion years, we can summarize:

for $B^2 = 10$ m^{-2} τ :   1.81 to 1.68 billion years

$B^2 = 15$ m^{-2} τ :   1.81 to 1.73 billion years

$B^2 = 20$ m^{-2} τ :   1.81 to 1.76 billion years

for loose water content V from 8% to 12%

and for uranium content 20% to 28%

and for poison content of about 50 ppm.

For higher poison contents these limits shrink down.

CHAPTER - III
STUDY OF SUSTENANCE OF REACTION

3.1 Dynamic model of reactors :

In this chapter a mathematical model of generation and burnup of different isotopes, when reaction at Oklo begins, has been made. This study has been motivated by the desire to investigate sustenance of chain reaction in Oklo Reactors. In the below developed formalism we would keep fluence as well as flux as open variables along with eight open parameters considered in criticality model. Following set of additional assumptions have been made

1. Time for which reactors operated is negligible compared to geological time. Hence geological time and reactor time have been treated as unlinked variables.

2. Reactors operated continuosuly without break. Or breaks were small enough so that natural decay of major isotopes can be neglected.

3. Flux remained constant all throughout reactor operation.

3.1.1 Burnup/buildup equations :

Once chain reaction begins burning up of U and initially present poisons effect K_{eff}. In the process various uranium and plutonium isotopes are formed, also formed are poisonous fission products. Isotopes which are formed in appreciable quantities such that they effect K_{eff} are U-236, Pu-239, Pu-240, Pu-241, Xe-135, Sm-149 and other fission products.

Out of them Pu-239 and Pu-241 are fissionable and contribute positively to K_{eff}.

Distinct feature of following isotope density equation is that fluence (Θ) as well as flux (φ), occurs in them in contrast to man made reactor burnup/buildup equation where only fluence is a variable. This is done to include the possibility that reaction operation period could be comparable to isotope decay half lives. Such a possibility has been suggested by the study of isotopic abundance in Oklo samples. In fact the lower limit to reaction operation period had been suggested to be 0.2 million years by the same study.

Burnup of U-235 can be described by following equation

$$\frac{dN_{25}}{d\Theta} = -N_{25}\,\sigma_a^{25} + \lambda_{29}\,N_{29}/\varphi - [\varepsilon\,P_1(1-p_{25})]\,x$$

$$(\,\bar{\nu}_5\,N_{25}\,\sigma_f^{25} + \bar{\nu}_9\,N_{29}\,\sigma_f^{25}\,) \quad (22)$$

Θ is fluence, φ is flux, λ_{29} is decay constant of Pu-239, $\bar{\nu}_5$ and $\bar{\nu}_9$ are neutron yield per fission of U-235 and Pu-239. First term takes care of loss of U-235 by thermal neutron absorption, second term gives gain in U-235 population because of Pu-239 decay. Third term represents loss of U-235 in epithermal region. P_1 is non-leakage probability till 180 Kev energy. Contribution of U-235 loss from neutrons born from Pu-239 fission is also considered in last term. Natural

decay of U-235 is ignored as its half life is of order of million of years where as reaction period order is most likely to be million hears. Following equation takes care of Pu-239 generation in Reactors.

$$\frac{dN_{29}}{d\theta} = N_{28} \, \sigma_c^{28} + [\epsilon p_1(1-p_{28})] \, (\, \bar{\nu}_5 N_{25} \sigma_f^{25} + \bar{\nu}_9 N_{29} \sigma_f^{29})$$

$$- N_{29} \sigma_a^{29} - \lambda_{29} N_{29}/\varphi \qquad (23)$$

The first two terms accounts for thermal and epithermal absorption in U-238 which leads to formation of Pu-239. Third term is loss of Pu-239 by absorption of thermal neutrons leading to fission or formation of Pu-240. The last term is isotope decay term which leads to loss of Pu-239 but gain in U-235 density. Epithermal and fast absorption by Pu-239 has been neglected.

Equation for Pu-240 is

$$\frac{dN_{20}}{d\theta} = N_{29} \, \sigma_c^{29} - N_{20} \, \sigma_a^{20} - \frac{\lambda_{20} \, N_{20}}{\varphi} \qquad (24)$$

First term in above equation is an capture component of third term in equation (23) and represents formation of Pu-240 by thermal neutron absorption by Pu-239. Second term gives loss because of thermal neutron absorption by Pu-240 which leads to formation of Pu-241 as fission cross section

of Pu-240 is negligible. Last term is loss because of isotopic decay of Pu-240. Here also epithermal/fast absorption is neglected.

Equation for Pu-241 is

$$\frac{dN_{21}}{d\theta} = N_{20}\,\sigma_a^{20} - N_{21}\,\sigma_a^{21} - \frac{\lambda_{21}\,N_{21}}{\cdot\,\eta} \qquad (25)$$

For every thermal neutron absorbed in Pu-240 one Pu-241 atom is born is what the first term means. Second term gives loss because of thermal absorption of neutron, part of these thermal absorption leads to fission. Third term is natural decay term. Here again epithermal/fast effect have been neglected.

U-236 is also formed in large quantities and its number density is constrained by following equation.

$$\frac{dN_{26}}{d\theta} = N_{25}\,\sigma_c^{25} - N_{26}\,\sigma_a^{26} \qquad (26)$$

First term is formation of U-236 from thermal neutron capture in U-235. Second term is burningup of U-236. U-236 formation by epithermal capture in U-235 has been neglected.

Most important burnable poisons are small quantities of rare earths and other elements with strong capture cross-section. Their presence had been idealised by B^{10} equivalent measure in Section

Equation for B^{10} number density is

$$\frac{dN_B}{d\Theta} = - \sigma_a^B \; N_B \qquad (27)$$

Only term present represent burn-up of poisons.

Fission of various isotopes (U-235, Pu-239, Pu-241) leads to formation of fission products which adds to parasitic capture of neutrons. Most important isotope build-up from capture point of view is of Xe-135 and Sm-149. Xe-135 is born in fission process as well as is formed as decay product of other fission products. Below given diagram illustrates isotope decay chain leading to formation of Xe-135.

$$Te^{135} \xrightarrow{\;\sim 1 \text{ min.}\;} I^{135} \xrightarrow{\;6.7 \text{ hrs.}\;} Xe^{135} \xrightarrow{\;9.2 \text{ hrs.}\;}$$

fission (on Te), fission (on I), fission (on Xe)

$$Cs^{135} \xrightarrow{\;2\text{x}10 \text{ yrs.}\;} Ba^{135} \; (\text{stable})$$
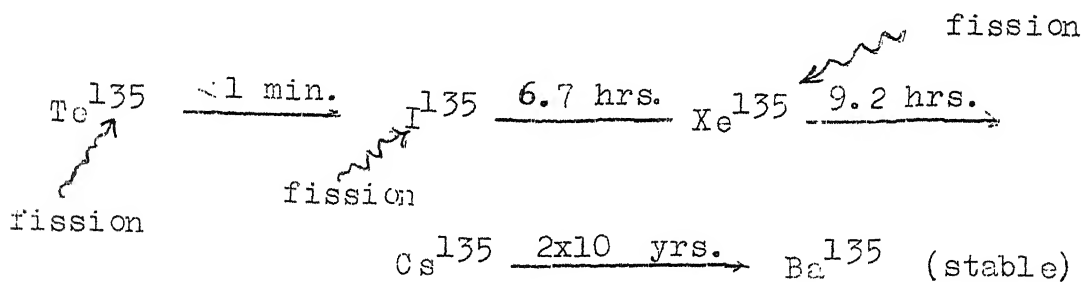
We can assume that all Te-135 formed immediately decays into Ir-135 and does not absorb neutrons to create 136 mass particle. To evaluate Xe-135 number density I-135 number density has to be known. Below given equation describes number density of I-135 isotope.

$$\frac{dN_I}{d\Theta} = -N_I \; \sigma_a^I \; -\lambda_I \; N_I \; /\varphi + (\gamma_5^I \; \sigma_f^{25} N_{25} + \gamma_9^I \sigma_f^{29} N_{29})$$

$$(28)$$

First and second term describe depletion of I^{135} by absorption and decay respectively. In last term γ_5^I and γ_9^I are fission yields of Iodine and Tellirium combinded, from U-235 and Pu-239 fission respectively. They have been multiplied by macroscopic fission cross-section of U-235 and Plutonium-239. Once NI^{135} is known from above equation it can be imbedded in Xe-135 equation given below to get number density of Xe-135.

$$\frac{dN_{Xe}}{d\theta} = - N_{Xe}\, \sigma_a^{Xe} - \lambda_{Xe}\, N_{Xe}/\varphi + \lambda_I\, N_I/\varphi$$

$$+ (\gamma_5^{Xe}\, \sigma_f^{25}\, N_{25} + \gamma_9^{Xe}\, \sigma_f^{29}\, N_{29}) \qquad (29)$$

First and second terms are decay and capture term of Xe-135. Third term is production of Xe-135 by decay of I-135. Last term is fission yield of U-135 and Pu-239, respectively.

Sm-149 is stable fission product. It is not only formed as fission product alone but also is a product decay of Pm-149 is

$$\frac{dN_{Pm}}{d\theta} = - N_{Pm}\, \sigma_a^{Pm} - \lambda_{Pm}\, N_{Pm}/\varphi + \gamma_5^{Pm}\, \sigma_f^{25}\, N_{25}$$

$$(30)$$

Pm-149 is not born in Pu-239 fission, hence third term only contains yield by U-235 fission. First and second terms are decay and neutron capture terms.

Equation for Sm-149 is

$$\frac{dN_{Sm}}{d\theta} = - \sigma_a^{Sm} N_{Sm} + \lambda_{Pm} N_{Pm}/\varphi + \gamma_9^{Sm} \sigma_f^{29} N_{29} \quad (31)$$

Sm-149 is not born in U-235 fission, hence last term only contains contribution from Pu-239 fission. First term represents neutron capture by Sm-149 leading to its depletion. Second term describes contribution from β decay of Pm-149.

Rest of the fission products can be considered to be equivalent to a hypothetical element with microscopic cross-section of 80 barns till 10^{14} fluence and 66 barns after that. The equation governing number density of fission products can be written as

$$\frac{dN_{f \cdot p}}{d\theta} = -\sigma_a^{f \cdot p} N_{f.p.} + 2(\gamma_5^{f \cdot p} \sigma_f^{25} N_{25} + \gamma_9^{f \cdot p} \sigma_f^{29} N_{29} + \sigma_f^{21} N_{21})$$

$$(32)$$

In equations (28, 29, 30, 31) contribution to the number density from fission of Pu-241 had been neglected. In equation (32) we have partially made up for it by including contribution from Pu-241 fission and taking cummulation yield of it's fission products to be one. The first term in above equation is an absorption term of an hypothetically unified fission products. Contribution from fast fission of U-238 and epithermal fission of other fissile isotopes have been neglected.

Number density of U-238 during reactor working can be described by following equation.

$$\frac{dN_{28}}{d\theta} = -\sigma_c^{28} N_{28} - [\varepsilon\, P_1 (1-p_{28}) + (\varepsilon-1)]\beta(\bar{\nu}_5 \sigma_f^{25} N_{25}$$

$$+ \bar{\nu}_9 \sigma_f^{29} N_{29} + \bar{\nu}_1 \sigma_f^{21} N_{21}) \qquad (33)$$

Here first term represents loss of U-238 by capture of thermal neutron. First term in square bracket gives loss because of epithermal capture and second term gives loss because of fast fission of U-238.

There are six other elements of importance present in reactors. They are SiO_2, Al_2O_3, FeO, MgO_2, K_2O, MnO. They will get depleted by capture of thermal as well as epithermal neutrons, though capture of epithermal neutron is quite less compared to thermal neutrons as is explicit from last chapter. Neglecting epithermal capture their number densities can be written as

$$N_r = N_r^o \exp(-\sigma_a^r \theta)$$

where N_r^o is number density of each of them at the start of reaction. As σ_a^r is of order of 10^{-24} fluence till $10^{21} - 10^{22}$ will make negligible effect on their number densities. Hence their depletion can be neglected if reactor behavior till about $10^{21} - 10^{22}$ fluence have to be studied.

Now we have twelve first order equation(22 to 33), some of them coupled, ,describing build-up and burn-up of crusial isotopes from criticality point of view.

3.1.2 *Temperature dependence of cross-sections :*

Cross sections and other data needed for solving above set of twelve equations is given in Appendix (I) for neutron velocity of 2,200 m/sec. The temperature dependence of needed cross-sections which do not obey 1/v law are as follows:

u-239:

$$\sigma_a^{29} = 1031.1(0.913+0.427 \ Tn/200)[293.4/(Tn+273)]^{0.5}/1.128 \text{ barns}$$

$$\sigma_f^{29} = 747.73(0.924+0.322 \ Tn/200)[293.4/(Tn+273)]^{0.5}/1.128 \text{ barns}$$

u-240:

$$\sigma_a^{20} = 295(1.017+0.065 \ Tn/200)[293.4/(Tn+273)]^{0.5}/1.128 \text{ barns}$$

e-135:

$$\sigma_a^{Xe} = [3.9753-(0.445/400)(Tn+273)] \ 10^6 \text{ barns}$$

1-149:

$$\sigma_a^{Sm} = [4.2(1.804+0.289 \ Tn/200)[293.4/(Tn+273)]^{0.5}/1.128]10^4 \text{barns}$$

Tn is neutron temperature in above equations. For rest of cross-section needed, $1/\sqrt{T}$ dependence on temperature was used. For hypothetically unified fission produts, absorption cross-section is a function of fluence and as no analytic form for it was available following descrete function can be used:

$$\sigma_a^{f.p.} = \begin{cases} 80[293.4/(Tn+273)]^{0.5}/1.128 & \text{for } 0 < \theta < 10^{15} \\ 66[293.4/(Tn+273)]^{0.5}/1.128 & \text{for } 10^{15} < \theta \end{cases}$$

3.1.3 Scheme for numerical solution :

From results of Chapter 2 we know conditions under which chain reaction would have began in Oklo. Once chain reaction began behavior of reactor can be known by embedding solution of above given set of twelve differential equations in criticality programme developed in chapter 2. Analytic solution of above given set of equations is problimatic. Scheme for numerical solution of above equations was developed using Runge-Kutta method which is an fourth order approximation method. Computer programme made for solution of these equations is given in Appendix (III) and was designed to be used as an subroutine for main programme of chapter 2.

As the solution of above given equations is needed over a very large range of fluence from zero to 10^{21} and order of different coefficients in these equations also is scattered very much lot of computational problems came up. Choise of step length became very important to minimise error. Dicisive advantage of

Runge-Kutta method is that control of step length from within
in possible so that error can be kept within desired limit.   In
Runge-Kutta method at every step four auxiliary quantities are
computed for every equation.   If there are m coupled equa-
tions like $y^{(i)\,\prime} = f(\Theta, y^{(1)}, y^{(2)},\dots y^{(m)})$, where $y^{(1)}$ to $y^{(m)}$
are m variables, than at n^{th} step of step length h, for i^{th}
equation these four quantities are

$$A_n^i = hf_i(\Theta_n, y_n^1,\dots,y_n^i,\dots,y_n^m)$$

$$B_n^i = hf_i(\Theta_n + \tfrac{1}{2}h, y_n^1 + \tfrac{1}{2}A_n^1,\dots,y_n^i + \tfrac{1}{2}A_n^i,\dots,y_n^m + \tfrac{1}{2}A_n^m)$$

$$C_n^i = hf_i(\Theta_n + \tfrac{1}{2}h, y_n^1 + \tfrac{1}{2}B_n^1,\dots,y_n^i + \tfrac{1}{2}B_n^i,\dots,y_n^m + \tfrac{1}{2}B_n^m)$$

$$D_n^i = hf_i(\Theta_n + h, y_n^1 + C_n^1,\dots, y_n^i + C_n^i,\dots, y_n^m + C_n^m)$$

And the new value of ith variable is

$$y_{n+1}^i = y_n^i + \tfrac{1}{6}(A_n^i + 2B_n^i + 2C_n^i + D_n^i)$$

In this scheme other than round-up error and truncation
error, an error is caused by the fact that slope f is evaluated
at $(\Theta_n, y_n^1,\dots, y_n^m)$ instead of at $(\Theta_n, y_n^1(\Theta_n,\dots),\dots,y_n^m(\Theta_n,\dots))$
This last kind of error poses computational problems, if absolute
value of any partial derivative $f_y = \partial f/\partial y$ is large this

error becomes significant. In our set of equations decay terms lead to this problem, which is absent in set of burnup/ buildup equations of man-made reactors. It can be shown that this error could be controled if step length h is chosen such that quantity

$$k_n^i = 2\left[\frac{C_n^i - B_n^i}{B_n^i - A_n^i} \right] \tag{34}$$

is within, say, $0.05 \leq k_n^i \leq 0.2$ for every equation. If any $k_n^i > 0.2$ then h must be decreased by say 50% and if $k_n^i < 0.05$ for all equations then h should be increased to say, double its value. This lower limit is to minimise number of steps to be computed. This ristriction on h brings it down increasing number of steps hence increasing round off errors. Round off error was minimised by working in double precision in which 16th digit is rounded of. Truncation error for every step in Runge-Kutta scheme is of the order of $[h/\theta]^5$ where θ is interval which has to be covered with steps of length h. Truncation error just gives upper tolerable limit, H, to step length. The value of h found from criterion of equation (34) should in no case go above H. With all these criterion error per step can not go above order of $[h/\theta]^5$ Another alternation check for accumulated absolute error order was followed. Equation for B^{10} equivalent poisons has simple exponential analytic solution. Result of analytic solution and numerical solution were matched for assessment of absolute roundup and truncation error

3.2 Results :

3.2.1 Fluence achieved after criticality :

A situation was chosen when reactor becomes just critical and for particular assumed flux, φ, duration till K_{eff} becomes less than one was computed using computational module developed for burnup study. Duration of reaction, T, for different assumed flux, φ, is plotted in Fig. (21). Both axes are on logarithmic scale. Fluence (= $\varphi \cdot T$) reached by reaction was seen to vary from 10^{17} to 10^{19} neutrons/cm^2, depending on flux. Fluence was less for lower values of flux. This is illustrated by the deviation of actual curve from dotted line which represents relation between T and φ for constant fluence of 10^{19} n/cm^2. Reason for this deviation is that for lower flux there is more natural decay of isotopes, in other words there is more Xe-135 and Sm-149 production as well as less number density of Pu-239 and Pu-241 for same fluence. All of these changes contribute negatively to neutron multiplication factor so that net fluence reached till K_{eff} becomes less than one is less. Number density of different isotopes of importance at fluence 10^{17} n/cm^2 for different values of flux is given in Table (7). Here we can see number density of Xe-135, Sm-149 and Pu-241 decreases with increase in value of flux. Xe-135 and Sm-149 number density decreases faster because of smaller decay constants of I-135, Xe-135, Pm-149 and Sm-149. Number density of Pu isotopes show insignificant change in the range of flux 10^{10} to 10^{19} neutrons/cm^2-sec. In this flux range change in Pu-239 and Pu-240 was
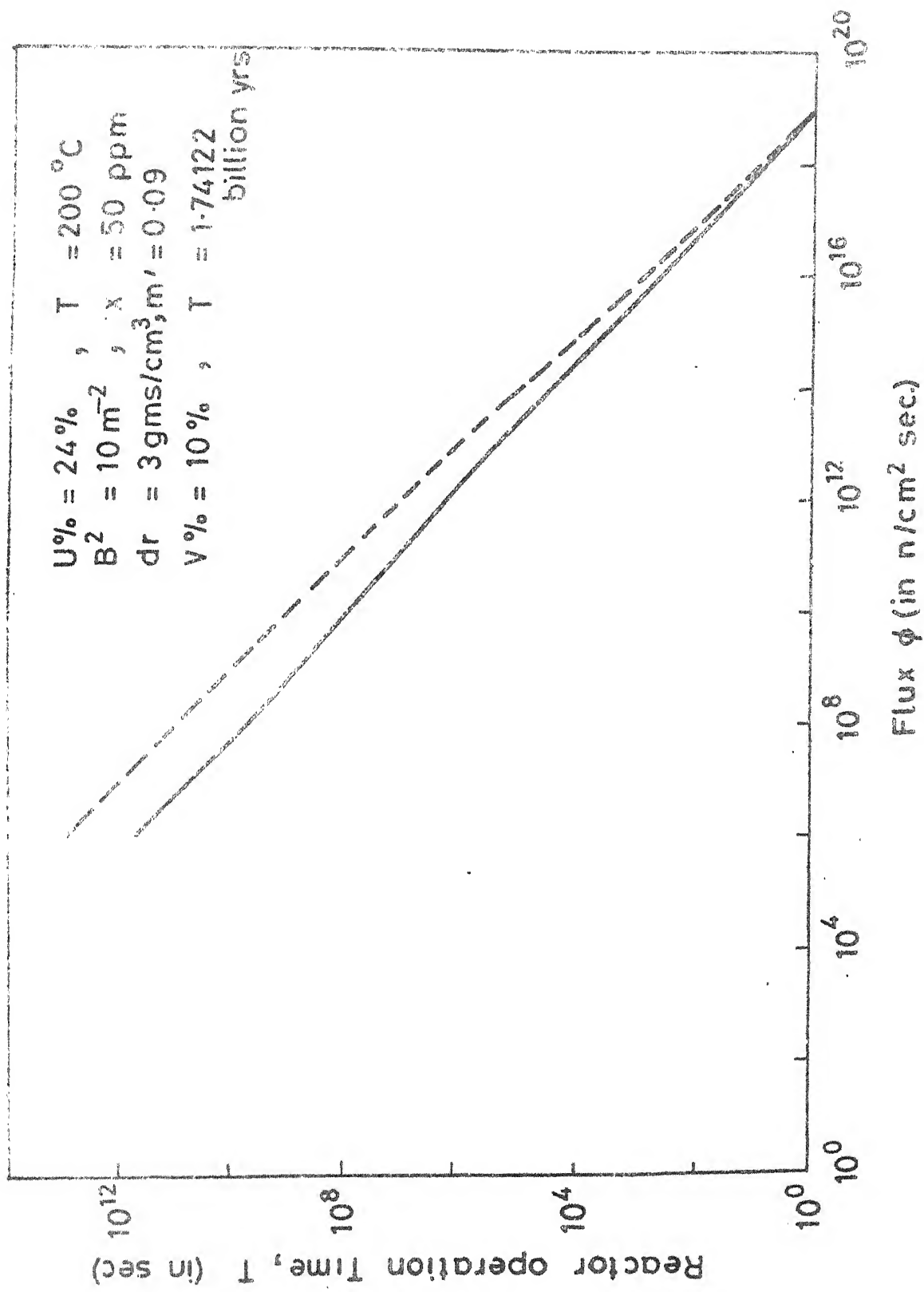
Fig. 21 Relation of the time till which Reactor operates without further accretion of Uranium with Neutron flux

TABLE 7

FLUX φ $n/cm^2/sec$	10^{10}	10^{14}	10^{18}	10^{19}
FLUENCE θ n/cm^2	10^{19}	10^{19}	10^{19}	10^{19}
N_{U-235}	0.182475×10^{21}	0.182475×10^{21}	0.182475×10^{21}	0.182475×10^{21}
N_{Pu-239}	0.35484×10^{16}	0.35484×10^{16}	0.35484×10^{16}	0.35484×10^{16}
N_{Pu-240}	0.2608×10^{11}	0.260836×10^{11}	0.260836×10^{11}	0.260836×10^{11}
N_{Pu-241}	0.80826×10^{5}	0.80228×10^{15}	0.8022825×10^{5}	0.8022825×10^{5}
N_{U-236}	0.92×10^{15}	0.92×10^{15}	0.92×10^{15}	0.92×10^{15}
N_{B}	0.312448×10^{18}	0.312448×10^{18}	0.312448×10^{18}	0.312448×10^{18}
N_{I-135}	0.1987×10^{13}	0.246426×10^{15}	0.247967×10^{15}	$0.24796723 \times 10^{15}$
N_{Xe-135}	0.271×10^{13}	0.2336×10^{13}	0.7979×10^{12}	0.79776×10^{12}
N_{Pm-149}	0.364×10^{13}	0.56865×10^{14}	0.56909×10^{14}	0.5691×10^{14}
N_{Sm-149}	0.532×10^{14}	0.453897×10^{11}	0.96757×10^{9}	0.96357×10^{9}
$N_{f.p.}$	0.752×10^{16}	0.752×10^{16}	0.752×10^{16}	0.752×10^{16}

U% = 24% , x = 50 ppm, B^2 = 10 m^{-2}, d_r = 3 gm/cm^3, m'=0.09,

V% = 10% , τ = 1.74122 billion years, T_m = 200°C.

Conversion ratio = 0.608846 is constant.

detected to be of order of 10^4 and 10^{-2}, respectively. This
is expected as order of Pu decay is about $(10^{-13} NPu/\varphi)$, which
is negligible compared to other terms in higher flux range but
becomes significant at lower flux range when reactor working
time becomes comparable to decay constant at Plutonium. In
Fig. (21) duration of reaction for flux lower than 10^6 n/cm^2-sec
is not plotted. Computation of these points needed extremely
large number of itterations to keep results in desired error limit.
This was because controlled step length of fluence was small
owing to increased significance of decay terms. Even to evaluate
duration of reaction for 10^6 n/cm^2 flux about 0.3×10^6 itterations
were needed to keep error in desired limit. For evaluation of
above given results error in criticality situation chosen is
equivalent to $U\% = (24 \pm 10^{-3})\%$.

J.2.2 Additional deposition needed for sustenance of reaction :

In last section it has been demonstrated for one likely
situation that fluence after starting of reactor does not exceed
10^{17} to 10^{19} n/cm^2 range. For reactors to have worked till
fluence of 10^{21} n/cm^2 as suggested by isotopic studies [42].
Some mechanism of compensating for loss of criticality is needed.
As was pointed out earlier situation of criticality was achieved
during desilicanisation process in which uranium got reconcentrated
in reaction zones. We hypothesis that after reaction began
desilicanisation process continued and uranium kept on depositing
in reaction zones. In this section we will give computed values

of quantity of additional mineralisation needed to sustain
reaction till fluence of 10^{21} n/cm^2 for different initial
criticality situations and assumed flux.

For criticality situation with U% = 24% additional
mineralisation needed to sustain reaction has been tabulated
in Table (8) for assumed flux of 10^8 n/cm^2 and 10^{19} n/cm^2.
Quantity of uranium added in reactor zone is represented by
increased U% (content of uranium by weight in percentage).
Here we note that for higher flux less additional mineralisation
is needed. This is because more Pu-239 and Pu-241 are produced
for higher flux leading to more fission. At fluence 10^{20} n/cm^2
Pu-239 and Pu-241 number density is 0.3413x10^{20} and 0.967x10^{17}
for flux 10^{14} n/cm^2-sec and is 0.864x10^{19} and 0.11x10^{13} n/cm^2
for flux = 10^{18} m/cm^2-sec.

In Oklo reactors only in some localised area uranium
content was as high as 60% [9]. Otherwise content of uranium
was scattered between 20% to 30% . The results in Table (8)
shows that even for additional mineralisation of 6% fluence
achieved is near 10^{20} n/cm^2. This indicates that initial
situation of 24% uranium could at the most be a localised
situation in reaction zone. For four more examplary situations
additional mineralisation needed have been computed for two
values of the 10^8 n/cm^2-sec and 10^{14} n/cm^2-sec and are given
in Table (9). Initial situation with lower U% were chosen to
allow larger addition of uranium. Two situation with high poison
content, x, were chosen to allow for help in sustenance by

TABLE 8

$U\% = 24\%$, $V\% = 10\%$, $x = 50$ ppm, $B^2 = 10 \ m^{-2}$

$T_m = 200^{\circ}C$, $d_r = 39 \ cm^3$, $m_1 = 0.09$, $\tau' = 1.74122$ byrs.

FLUX	10^8	10^{14}
Fluence at which mineralisation is resumed.	0.7026×10^{18}	0.13×10^{19}
FLUENCE	U%	U%
0.1×10^{20}	25.25%	24.25%
0.25×10^{20}	26.30%	24.59%
0.5×10^{20}	27.38%	26.25%
0.75×10^{20}	28.14%	25.9 %
0.1×10^{21}	28.84%	26.62%
0.25×10^{21}	32.90%	30.82%
0.5×10^{21}	39.04%	37.76%
0.75×10^{21}	45.68%	44.35%
0.1×10^{22}	51.71%	50.30%

TABLE 9

Group parameters

- (a) $U\% = 20\%$, $V\% = 10\%$, $x = 40C$ ppm, $B^2 = 10\ m^{-2}$, $\tau = 1.9$ billion years
- (b) $U\% = 20\%$, $V\% = 10\%$, $x = 50$ ppm, $\tau = 1.786$ billion years, $B^2 = 10\ m^{-2}$
- (c) $U\% = 15\%$, $V\% = 10\%$, $x = 50$ ppm, $B^2 = 10\ m^{-2}$, $\tau = 1.8866$ by
- (d) $U\% = 16\%$, $V\% = 12\%$, $x = 40C$ ppm, $B^2 = 20\ m^{-2}$, $\tau = 1.97$ by

	(a)		(b)		(c)		(d)	
FLUX	10^{8}	10^{14}	10^{8}	10^{14}	10^{8}	10^{14}	10^{8}	10^{14}
Fluence after which mineralisation starts	0.5×10^{17}	0.1×10^{18}	0.2×10^{16}	0.2×10^{16}	0.2×10^{14}	0.2×10^{16}	0.2×10^{16}	0.2×10^{16}
FLUENCE in n/cm²	U%	U%	U%	U%	U%	U%	U%	U%
0.1×10^{19}	20.177	20.18	20.09	20.18	16.55	16.09	16.18	16.09
0.563×10^{19}	—	—	—	16.91	16.09	16.09	16.09	16.09
0.316×10^{19}	—	—	20.53	—	16.55	16.55	16.28	16.28
0.178×10^{19}	—	20.35	20.01	16.09	16.09	—	—	16.09
0.1×10^{20}	21.13	20.18	20.26	17.72	16.37	17.095	17.63	16.18
0.563×10^{20}	—	20.79	20.18	17.18	16.18	16.09	—	16.09
0.316×10^{20}	—	20.35	20.01	16.91	16.55	16.46	—	16.46
0.178×10^{20}	21.90	20.18	20.53	18.34	16.55	17.63	17.63	16.28
0.1×10^{21}	23.48	23.72	20.61	19.21	17.01	18.25	18.36	16.92
0.563×10^{21}	33.35	31.455	37.61	35.87	35.46	32.59	30.15	28.87
0.316×10^{21}	27.81	25.46	31.25	29.04	28.53	25.74	—	22.44
0.178×10^{21}	24.92	22.57	27.29	25.00	24.24	21.47	—	19.21
0.1×10^{22}	42.66	42.41	47.07	46.82	45.56	43.46	41.42	40.04

burning up of initially present poisons. Also a situation was chosen with high poison content and smaller size of assembly to accomodate possibility that reaction could have began in smaller area which is capable of susteinance as well as can feed neutrons to the rest. From Table (1) we know that age of primary mineralisation falls around 1.9 billion years. Considering results of Chapter 2 we can conclude that it is unlikely for Oklo reactors to have become critical for U% less than 16%. Two situations for U% = 16% and other two for U% = 20% were taken.

In none of the situations in Table (9) fluence would have reached 10^{21} n/cm^2 within constraint on uranium content infered from fossils of Oklo reactors. Fluence reaching 10^{20} n/cm^2 is possible with 2.5% to 6% additional mineralisation for initial uranium content of 15% to 24% , V% = 10%, poison content of 50 ppm and flux range 10^8 n/cm^2-sec to 10^{14} n/cm^2-sec. If neutron flux in reactors was less than 10^8 n/cm^2-sec than this range shifts to higher values and shifts to lower values for higher flux. Another point to note here is that additional mineralisation needed is less for higher initial uranium content. Considering constraint on uranium content of Oklo reactor fluence till 0.3×10^{21} seems possible (in situation (c)). For higher poison content fluence reachable in reactors increases. Situation (a) and (d) have extremely high poison content. From (a) and (d) we can roughly draw upper limit to fluence reached as around 0.5×10^{21} n/cm^2. This needs 8% to 12% extra mineralisation with initial uranium content between 16% to 20% . We can

conclude that in most of the reaction area fluence between 10^{20} n/cm^2 to 5×10^{20} n/cm^2 would have been reached with at least 2.5% to 6% additional uranium for initial uranium content between 16% to 24% and poison content of 50 ppm or more.

Some localised areas in reaction zones have been reported to have had more than 30% uranium content. In reaction zone 2 fairly large area had uranium concentration upto 40%, at some spots in it concentration is 60%. In these locations there is a possibility that larger value of fluences would have been reached.

4.1 On general possibility of nuclear chain reaction in Nature :

Apparently the only major roles of the fission process in nature are to provide an upper limit to mass number and to some extent influence the abundance of fission products. Apart from this we may forget about fission if we are interested only in the major features of our world. The fact that the number of neutrons emitted per fission, $\bar{\nu}$, is around two seems to be of very minor importance to nature. Also there is no pressing theoretical reason that $\bar{\nu}$ be greater than one. In fact one of the earliest nuclear researcher Victor Wiesskopf reflecting on pre-second world war days had written " Many of us hoped that the number of neutron per fission would be low enough to prevent making of a bomb" [52]. But $\bar{\nu}$ is large enough for nuclear chain reaction to be possible. Now we know at least in Oklo nature had made use of this possibility. Other possibility of occurance of nuclear chain reaction in nature are there. This is in the period following formation of earth. One school of historians of earth feel that earth was cooler when it first formed because Neon is extremely rare in our atmosphere but comparably abundant in stars. Neon's weight is same as molecular weight of water and water differs from Neon chemically in that it reacts under 'cool' conditions to form compounds where as Neon is inert. This would mean rejection of hypothesis that earth's mental is magma. In the process of formation of

earth from plasma cloud most of the uranium would have been
bracketed between iron core and surface because of gravitationa
sorting and ionisation energy differenciation. In the process
of crystallisation of rocks under pressure uranium rich fluid
would have been expelled [9]. As U-235 enrichment at the time
of formation of earth 4.5 billion years ago was 23% relativel;
small mass of uranium would have gone critical. But fossiles
of these reactors, if at all they exist, would be still buried
deep under crust of earth. Unless some orogeny related phenomer
has led to their exposure there is little possibility of detecti
them. On surface of earth till the end of Hedean period 3.4
billion years ago uranium remained as disseminated UO_2 in cryst
lline rocks as there were no known mechanisms which could have
concentrated uranium. During Archean period (about 3.4 to 2.6
billion years ago) procaryotic life forms evolved but there was
no significant amount of free oxygen during this time and uraniu
remained as unoxidised UO_2. In this period uranium did concentr
by means of gravitational sorting of heavy stream sediments, whi
included UO_2. These unoxidised ores known as paleoplacer or
quartz-pebble conglomerate ore rarely contained more than few
tenths % uranium and even though relative U-235 content was 15%
to 8% there was total insufficient uranium concentration for
criticality. Then during Proterozoic (about 2.6 to 2.0 billion
years) important changes occured. This was the period of intens
mountain building and subsequent weathering. Evolution of
eucaryotic life capable of photosynthesis librated oxygen and an
extensive surface 'reservoirs' of unoxidised uranium bearing

sediments and orogeny related exposures were attacked by oxygen and large quantity of complexed uranyl ion were hydrochemically released. This uranyl precipitated as uraninite, pitchblende, or coffinite often resulting in large massive views of uranium. Theyrrepresent world's richest reservoirs. Though U-235 concentration was 4% to 2% reconcentration of uranium because of tectonic activity could led to criticality. Oklo reactors evolved in this period and there is no special reason why many more such reactors would not have worked in that period. Next major reconcentration of uranium occured when plants evolved. The organic debris of plants in terrestrial sediments lead to formation of pitchblende vein deposits. But it was too late. Concentration of U-235 had dropped to a level when chain reaction was possible only in some specific geometries. Several years ago some petrified trees were discovered in the Colorado mine in the vicinity of the San Migual River deposit. One of the trunk was about 30 meters long and a more than a meter thick and other a little less. Over a hundred tons of uranium ore was obtained from these trunks ! From this amount of ore some six tons of UO_2, about 2 gms of radium and number of other elements were extracted [53]. Had there been trees in proterozoic period there would have been thousands of Natural nuclear Reactors.

4.2 Conclusions :

(1)   Effective neutron multiplication factor, K_{eff}, does not have a monolithic dependence over temperature. Till about temperature of $240^{\circ}C$ temperature coefficient of reactivity

positive after that it becomes negative (from Sec. 2.3.3).
This will have important implication for deciding flux level
reached in reactors. Flux is dependent on excess reactivity
which in case of Oklo reactors is because of rise in temperature.
If initial temperature of milieu was more than the temperature
at which K_{eff} is maximum then reactors would have worked at
very low flux.

(2)   Slowing down length and thermal diffusion length are
        considerably small in reaction zone (from Sec. 2.3.4). If
        outside reactor zone thermal diffusion length is small,
        which is likely to be the case, we can say that neutrons
        from our reaction zone did not reach other in appreciable
        quantity. This wouldindicate that reaction zone 3-4 and
        5-6 (in Fig. 6) were not coupled reactor systems. Such
        possibility was hinted by Maurett [9].

(3)   In Oklo reactors leakage was very less,   1%., because
        of its unique moderation feature and high parasitic capture.
        This has a massage for radiation safety concern. Average
        logarithmic energy decrement, number of collisions needed
        for slowing down and slowing down time of Oklo moderator is
        slightly better than that of D_2O moderator but moderating
        power is much better than even that of natural water.
        This is because of high number density of Oklo moderator.
        On the other hand parasitic capture in thermal as well as
        epithermal region is quite high. It is as if Oklo moderator
        absorped neutrons which were to have leaked out. This points

to an new reactor concept in which leakage is quite less at the same time it serves a purpose of production of useful isotopes.

(4)   Following set of conclusions can be drawn about conditi[on] under which Oklo reactors became critical :

(a) Criticality in Oklo was possible before 1.67 billion years. This lower limit on geological time is much less than age of primary mineralisation.

(b) Volume fraction of loose water (V%) greater than 6% is necessary for criticality. In other words volume fraction of microfractures in which water could stay should be more than 6% for criticality to be possible. Criticality condition in general was found sensitive to water content in reactors.

(c) Most likely reaction did'nt begin before 16% uranium content by weight had been accumulated in the process of desilicanisation.

(d) For all six reactors to have worked simultaneously either uranium content (U%) in smaller zones should have been 4% more than larger zones or loose water content should have been more in smaller zones.

(e) For loose water content 8% to 12% and uranium content 20% to 28% and poison content of 50 ppm.

for $B^2 = 10 \text{ m}^{-2}$ $\tau > 1.68$ billion years
$B^2 = 15 \text{ m}^{-2}$ $\tau > 1.73$ billion years

$$B^2 = 20 \ m^{-2} \qquad \tau \quad \sim 1.76 \ \text{billion years}$$

The upper limit is left open because of uncertainty in age of primary mineralisation.

(5)   Once reaction begins functioning fluence between 10^{17} n/cm^2 to 10^{19} n/cm^2 would have been reached without further accretion of uranium.

(6)   In most of the reaction area fluence between 10^{20} n/cm^2 to 5×10^{20} n/cm^2 would have been reached with at least 2.5% to 6% additional uranium for initial uranium content between 16% to 24% and poison content of 50 ppm or more. Fluence attained is sensitive to poisons. Maximum fluence reached through continued mineralisation considering bound on content of uranium inferred from fossiles of Oklo reactors is half of 10^{21} n/cm^2. Study of isotopic content of Th, Bi, Ru and Nd [41] suggests final fluence to be 10^{21} n/cm^2. In that case hypothesis of continued mineralisation alone can not explain sustenance of reaction.

4.3 Shortcomings of this study :

Major drawback of this study is that thermal and epithermal flux are not differentiated. In other words epithermal flux is exaggerated. This leads to overestimation of epithermal quantities. Another weak point in criticality model is calculation of fast fission factor for which very simplified formalism is used.

Another limitation of this study is that spacial distribution of different elements in reaction zone has been ignored. As far as study of criticality conditions and reactor physics features of reactors are concerned this is not much of a handicap but study of sustenance of reaction needs consideration of spacial distribution. If initially present poisons were distributed in such a way that near boundaries of reactor their content was much higher than at centre than reaction would have sustained for longer time as well as larger fluence. Such situations coupled with continued mineralisation could probably lead to achievement of even 10^{21} n/cm^2 fluence.

4.4 *Recommendations for further study :*

(1)   Kinetic aspects of Oklo reactors should be modelled and embeded into criticality and burnup/buildup scheme developed in this study. This makes flux as an determinate quantity. If this is done we will have a closed loop with K_{eff} determining flux and flux determining temperature and temperature inturn changing K_{eff}. For modelling of this closed cycle, along with reactor kinetic equations heat transfer properties of Oklo rock will have to be idealised. If all these additions can be done we can simulate Oklo reactors on computer. With this other than modifying results of sustenance of reaction mode of reaction can be studied.

(2) As was pointed out in Sec. 4.2 Oklo moderator has
interesting features. If points to new concept of safe
reactor in which neutron leakage is less and instead it
could produce useful radioisotopes. It might be an useful
exercise to develop this concept of safe reactor. An
extensive study can be done to find out possible and useful
moderating material along these lines. The search for
such moderator will depend on selection of elements which
can chemically form dense solid and have high moderating
power. Choice of these elements can be such that their
isotopes can be used for numerous radioisotope applications.

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1.1 Nuclear properties of materials.

Element or Compound	Atomic or Molecular weight	Density (gm/cm³)	$1-\mu_0$ (1)	Average logarithmic Energy decrement (1)	Thermal Microscopic cross-sections** (borns)			Epithermal Property	
					σ_a	σ_s	σ_{tr} (3)	σ_s/ep (borns)	Infinite dilution resonance Integral
H₂O	18.016	1.00	0.676	0.948	0.66	103	70.288	25.4144	0.5
Si	28.09	2.42	0.9762	0.0698	0.16	1.7		2.2	0.5
Al	26.98	2.699	0.9754	0.0723	0.241	1.4		1.4	0.18
Fe	55.85	7.86	0.9881	0.0353	2.62	11		11.4	2.3
Mg	24.32	1.74	0.9722	0.0811	0.059	3.6		3.4	0.9
K	39.1	0.87	0.9829	0.0504	2.07	2.07		2.1	3.5
Mn	54.94	7.2	0.9878	0.0359	13.2	13.2		1.9	15.4
O	16.00		0.95833	0.1209	≤0.0002	4.23	3813.0	3.75	0.18
B10									
U 235	235								
U 238	238.07	18.9	0.9972	0.0084		8.3		8.3	280

Contd...Appendix-I

1.2 Thermal data for isotopes of Uranium and Plutonium.

	U^{235}	U^{236}	U^{238}	Pu^{239}	Pu^{240}	Pu^{241}
σ_a	658.8	7.0	2.7	1031.1	300	1400
σ_f	549.1	--	--	747.73	--	1025
$\overline{\nu}$	2.47±0.03	--	--	2.49±0.01	--	3.0

1.3 Thermal data for fission products:

		I^{135}	Xe^{135}	Pm^{149}	Sm^{149}	f.p.
σ_a (barns)					4.2×10^6	80
Cumulative Fission Yield	γ^5 from U^{235}	0.061	0.0002	0.014	--	0.923
	γ^9 from Pu^{239}	0.058	0.0506	--	0.0189	0.8625

Contd...Appendix-I

1.4 Radioactive decay constants:

	Pu^{29}	Pu^{20}	Pu^{21}	I^{135}	Xe^{135}	Pm^{149}
	9.03273×10^{-13}	3.37×10^{-12}	1.67×10^{-9}	2.874×10^{-5}	2.11×10^{-5}	$3.6 \times 10^{-}$

1.5 Additional data used :

$$\eta_{25} = 1.7$$

※   Value is of infinite resonance absorption integral for U-235.

(1) Data is taken from ANL-5800.

(2) Data taken from Table (18), 2-13, Nuclear Engineering
     Handbook edited by Harold Etherington.

(3) Miscroscopic transport cross-section for elements other than
     water were calculated by following equation

$$\sigma_{tv} = \sigma_a + (1 - \bar{\mu}_o) \sigma_s$$

※※   All the cross-sections at neutron speed of 2200 m/sec.

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